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Particulate matter in and from agriculture

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Particulate Matter in and from Agriculture

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T. Hinz [1]

Introduction

Agriculture and the environment are connected and interacting in a more sophisticated way than normally worked out in the chain emission-transmission to exposure and effects. Agriculture is source of various materials which may affect all compartments of the environment - air, water, soil, plants and individuals. But agriculture is acceptor of many pollutants from different sources of the environment, too. At least agricultural areas are the environment e.g. for animals or human beings which use the fields, forests and fruit gardens for resting and recreation.

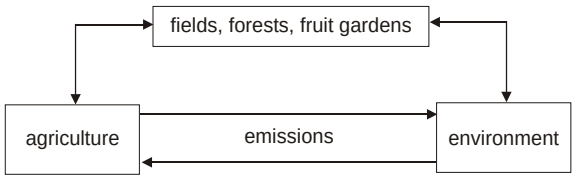

Figure 1:
Interaction between agriculture and environment

In the past mainly ammonia emissions and means for reduction have been discussed on nearly all levels of research, administration and legislation. Particulate matter (PM) - aerosols and dust - had been seen as a more secondary problem for health and welfare of man and animal. But newer more comprehensive knowledge shows that the effects of particles on individuals had been underestimated. Therefore limitations of PM found entrance into national and European regulations /1/.

In the following, sources and effects, transport models and measurements of PM in and from agriculture will be discussed under consideration of particle size and the composition of the sometimes very complex bio-aerosols.

Keywords: particulate matter (PM), bioaerosol, sources, emission, air quality, measurements

Definitions

Particles in air pollution and control require definitions concerning size and material which must be considered for measurements, transport and effects.

In the past particle size was distinguished as total dust or the respirable fraction according to the convention of Johannisburg /2/. Meanwhile new definitions are valid e.g. ISO 7708 defines health related particle size fractions, cf. figure 2 /3/. Inhalable, thoracic and respirable fractions are derived from the depth of entrance into the human breathing tract. Concerning ambient air US EPA defined PM10 and PM2.5 /4/. PM means particulate matter and the index numbers 10 and 2.5 are the cut-off diameters of particle separators that the total suspended particulates (TSP) have to pass. PM definitions are quite close to the health related fractions. Looking to the effects, ultrafine particles PM0.1 become more and more important.

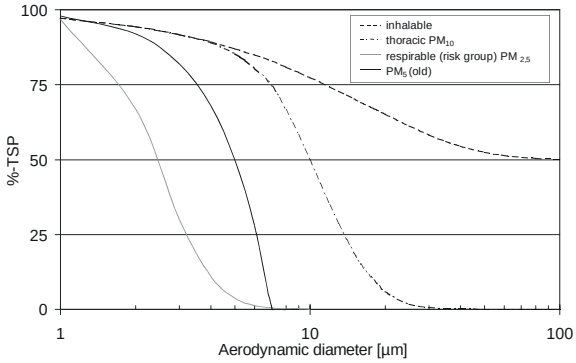

Figure 2:
Fractions of particles acc. to EN ISO 7708

The definitions above are only particle size related without any consideration of substances and materials. Particle dispersions from agriculture are composed by various matter of different origin. These bioaerosols consist of anorganic matter e.g. from soil, organic matter from plants or animals as well as dead and alive microorganism like germs, fungi, viruses, bacteria or parts of these e.g. endotoxins. This definition of bioaerosols doesn't reflect to particle size in a particular way.

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Sources

Certain operations and the use of materials like fuels, fertilizer, pesticides and animals' food and litter result in the emission of dusts inside buildings and outside in fields. In contrast to other commercial plants, emissions from agriculture are discontinuous with different frames of time. In plant production, the emissions depend on the stage of the fruit - seeding, growing, harvesting and soil cultivation. In animal production, the life cycle and kind of production of the different species has to be considered – dairy cows, fattening pigs, broiler and laying hens.

Figure 3 shows possible anthropogenic sources of dust produced by agriculture in plant (food and non-food) and animal production. There are stationary and mobile sources. Examples of stationary plant production systems are systems for cleaning and drying and for unloading, sorting and sacking the raw and final products. In animal production, the animals themselves produce particulates of organic materials e.g. from skin and feathers. Other sources of dust inside an animal house are the result of feeding, stocking and manure management. Emissions caused by combustion for heating the animal house can be calculated by specific emission factors and the consumption. NH_3 emissions are the origin of secondary fine particles, which may be part of long range transport /5/.

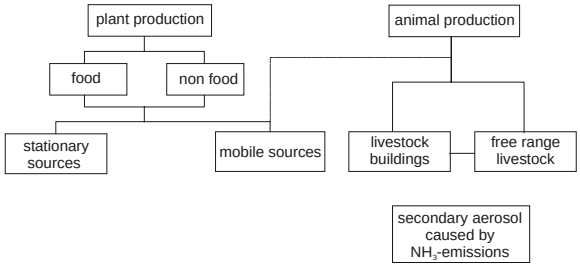

Figure 3:
PM-Sources of agriculture

Most mobile sources are associated with plant production although the transport and the application of organic fertilizers may be seen as part of animal production. Self-propelled farm vehicles e.g. combine – harvesters and tractors with mounted machines for soil cultivation, harvesting or spreading fertilizer or pesticides produce emissions of dusts in the fields. Particle emissions from the engines' exhaust can be calculated by fuel consumption and specific emission factors which depend on the kind of fuel and engine. In the international szenario of air pollution and control e.g. LRTAP (Longe Range Transboundary Air Pollution) these emissions are not considered specifically as part of agriculture but will be mentioned in the sectors of on and off road traffic /6/. Nevertheless these problems are subjects of agricultural engineering research /7/. Relevant sources agreed for consideration in the UN ECE Task Force for Emission Inventory and Projections are given in table 1.

Table 1:
PM sources of agriculture – UNECE-Activities

Plant production	**Feed management on the farm**	**Housed Livestock**
Harvesting	Milling	Dairy cows
Post-Harvest treatment	Mixing	Other cattle
Soil cultivation	Blowing	Fattening pigs (incl. Weaners)
Fertilizer application	Conveying	Sows
	Delivering in troughs	Ovines
	Handling the raw material	Horses
	Pellet-Making	Laying hens
	Blowing of materials delivered to the farm	Broilers
		Other poultry
		Fur animals
On field burning of stubble, straw …	**Nature**	**Free range livestock**

Measuring techniques

Depending on the task PM measurements require different instrumentation. PM emissions of forced-ventilated livestock buildings or capsulated plants for drying and cleaning cereals or greenfodder require isokinetic samples mostly combined with the gravimetric method according to VDI 2066 /8/.

Aim of isokinetic sampling is to get a representative collection of TSP. If information about special fractions are wanted e.g. for estimation of the content of fine particles for dispersion models or to design particle separators for emission reduction, particle size analysis is carried out e.g. using the Coulter Counter Technique or directly measured using a pre-separator with a known fractional collection efficiency curve.

For measuring air quality inside buildings a discontinuous gravimetric method is used if further analyses of dust material are wanted e.g. for microorganism and endotoxins. A quasi-online dust monitor is used if short time events must be detected over a day.

Health related samples are taken according to the convention of Johannisburg in which the respirable fraction is calculated the following penetration function:

$$P = 1-(d/d_0)^2 \quad d_0=7.07\mu m \quad \text{aerodynamic diameter}$$

Depending on the new conventions and regulations other particle size sensitive measuring techniques are developed. In principle, the following different ways of measurement are possible, cf. table 2.

Optical instrumentation is particle size sensitive but difficulties arise due to the composition of agricultural dusts. They are not homogenous in physical or optical density and are heterogeneous in shape as shown in figure 4 for a typical dust from a poultry house.

Figure 4:
Photomicrograph of dust from a poultry house, showing its heterogeneous nature

Thus optical instruments must be calibrated for the particular dust which is to be measured. The gravimetric method is used to provide the reference for the calibration /9/. Figure 5 shows the reading of an optical instrumentation versus dust concentration in a barn.

Table 2:
Particle size-selective measurements

Aerodynamic separation	Conventional filter weighing
	Online electronic balance
	Online optical detection
Sampling total dust	Online size selective optical detection
	Conventional filter weighing with additional particle size analysis

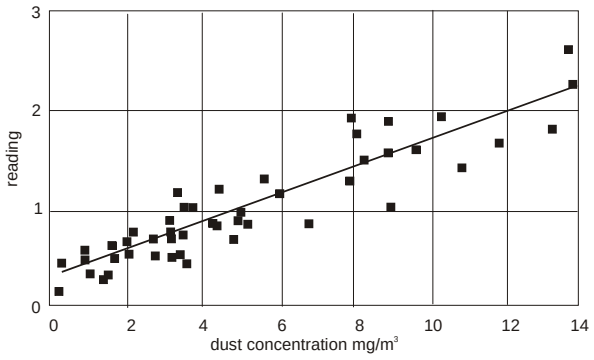

Figure 5:
Calibration curve of a Tyndallometer used in a Louisiana-type broiler house

Using total PM samples for additional particle size analysis may lead to failures in calculating the different fractions, if filters must be solved as necessary for the Coulter Counter. Agglomerates which are suspended in the air will be destroyed and a higher content of fine particles will be pretended. This effect is more severe measuring PM in livestock houses with high air humidity than measuring e.g. during combine harvesting or drying and cleaning cereals.

At the present time methods with aerodynamic separation are preferred.

Results

Cereal Production

During cereal unloading, cleaning and drying, dust is set free and emitted to the environment. 400 mg/m³ may be measured in the neighbourhood of the source during the short time of unloading. Because of the relatively coarse particles, the initial value of concentration decreases to 2 % at a distance of about 15 m.

For a drier with a capacity of 2-2,5 t/h, cereal exhaust concentrations between 1 mg/m³ and 100 mg/m³ with resulting mass flows (emissions) of 0,01 kg/h up to more than 0,6 kg/h have been measured. For a cereal cleaner with 20 t/h resp. 60 t/h the corresponding values are 17 mg/m³, 750 mg/m³ and 0,1 kg/h up to 5 kg/h. In the latter case dust separation is required. Particle size distribution of the emissions during drying shows mainly a higher content of fine particles than during cleaning. Up to 50% at drying but only 15% at cleaning had been measured /10/.

Both in drying and cleaning, the level of dust emission is strongly influenced by the kind of crop. Dust production from barley is more significant than from wheat.

Among mobile sources in plant production, combine-harvesters produce high dust emissions which also depend on the kind of crop, the soil, the climate and other parameters. Dust emissions are roughly proportional to the cut width /11/. In a particular case a width of 2 m resulted in emissions at a rate of 20 kg/h, while 4 m cut width resulted in 40 kg/h. Dust emissions at rates in excess of 60 kg/h have been measured. For creating emission inventories product related specific emission factors are required. For combine harvesting this factor is calculated in the range of 1.2-6.1 kg/t (TSP related to harvested cereal). Particle size distributions show aerodynamic 50 % diameters of about 15 µm and a potential of PM2.5 material mostly below 5 %.

Dusts in the neighbourhood of a combine harvester may cause allergic reactions. To protect the farmer against the airborne contaminants during mobile works in the fields a forced-ventilated, air-conditioned cab is the preferable way /12/.

Animal production

Dust from animal production inside livestock buildings is part of air quality and may affect the health and welfare of man and animal. Upon leaving the barn in ventilation flow or through other openings, dust becomes an environmental problem. Particles originate from the animal itself and from fodder and litter. They form a multi-component biological aerosol with a great variety of inorganic and organic material, because gases, bacteria and other germs are adsorbed and fixed on the surface of the dust particles. The strength of each source depends on parameters such as the species, stocking density, age of the animals, feeding and littering strategies, the climate conditions inside the barn and the behaviour of the animals. Environmental enrichments and e.g. lighting strategy influence the animals' activity and consequently dust emissions /13/. For international szenario, specific emission factors averaged for one year must be calculated. Therefore all time dependencies must be known on the scale of a day, the fattening periods and the season.

Dust emissions from cattle, pig and poultry houses in Northern Europe were measured and measurements of the air quality inside turkey houses were made, too.

Table 3 shows the results. The emission results are a part of a European Research Project /14/.

Table 3:
Mean emission rates of total dust on a 500 kg liveweight basis for different species

	Emission rates in mg/h (500 kg)		
	cattle	pig	poultry
Germany	184	651	2118
Denmark	128	1102	3509
Netherlands	143	674	3640
U. K.	97	633	3138

For the emission rates, the measured mass flow from a barn is related to the livestock unit (500 kg) basis, so that a direct comparison between the different species is possible. The lowest specific emission rates are found in cattle houses. The highest values are observed in poultry buildings especially for broilers on litter.

The values in the table are averages of measurements carried out at different times of the year in different barns in different countries. The aim of the European study was to provide the Commission with fundamental data regarding emissions in Northern Europe caused by animal production. Particle sizing of the emissions was not requested at that time.

For the farmer, more detailed information about the dust concentration in the course of a day may be more interesting, e. g. as a factor in animal health, welfare and productivity. Figure 5 shows the course of the total dust concentration in an experimental turkey house with a stock of 540 birds. Day and night time are to be distinguished significantly with higher value during the day. An averaged ratio can be

calculated to ~ 2. Day and night time can be influenced by the farmer by switching light on/off which is clearly detectable in the dust concentration, see figure 6.

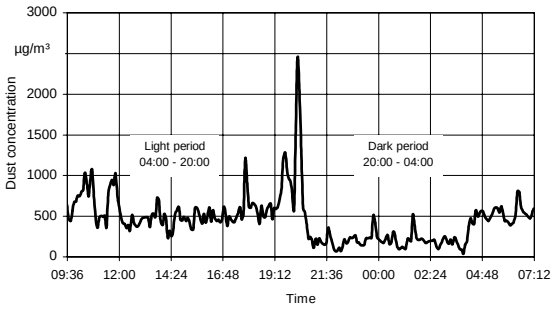

Figure 6:
Influence of lightening on dust concentration in a turkey barn

Measurements in another commercial freely ventilated and illuminated turkey production facility with a stock of 3000 birds show a proportion of 5 % PM 2.5 which means an average of 0.2 mg/m³ over a duration of 3h on day time.

The content of PM10 or PM2.5 depends on factors of production and the species itself. A previous study of the air quality and the emissions from a large piggery with wet feeding showed PM2.5 ranging between 40 and 70 %. For particle size analysis Coulter Counter Technique was used which may lead to a higher content of small particles than aerodynamic measures, as mentioned above. These values must be seen with the total emission flow and the specific emission factor which is calculated to be approximately 12g/(h animal). The exhaust concentration of PM2.5 concerned is in the order of 0.5-0.9 mg/m³ with flow rates of 4.8 – 8.4 g/(h animal) /15/.

As mentioned above PM emissions in and from agriculture - especially in animal production - have a high content of organic material inclusive germs. In normal air germs are found in concentrations of 10^3 to 10^4 per m³. In animal production germ concentration - bacteria and fungi - is 2 decimal powers higher than in normal air /16/. In figure 7 the most important fungi and bacteria of animal production are given. The lengths of the columns are a measure for their frequency. The most common species of fungi are scopulariopses and of the bacteria the various bacilli. It is conspicuous that staphylococci and streptococci are found. It must be mentioned that there is a wide spectrum of fungi in poultry production in contrast to the low variety in pig houses. This observation is valid for the bacteriological examinations, too.

Conclusion

PM concentration, PM emissions and emission factors must be related to the different definitions of size

- looking to human (animal) health and welfare
- looking to transportation and effects in the ambient air
- the complex composition of the aerosols in agricultural production requires sensitive use of given size selective measuring procedures and special attention to the content of microorganisms of parts of it – bioaerosols.

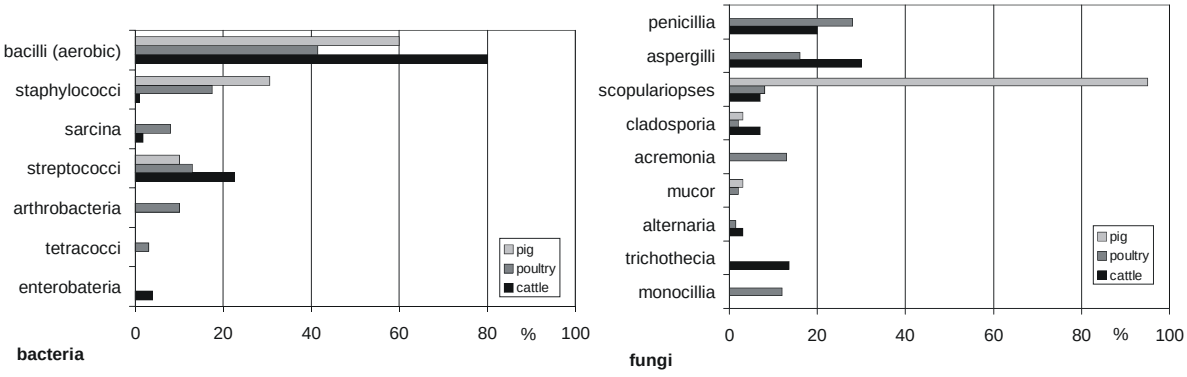

Figure 7:
Germ spectrum of total dust samples of animal production

Summary

Agriculture and the environment are connected interactively which means they emit and receive contaminants from each other, e.g. PM.

Emissions of agricultural PM result from animal houses, and the mobile and stationary equipment used in plant production. In comparison with other sources special time functions are to consider according to biological dependencies.

In most cases, PM emissions in and from agriculture affect man and animals in the living and working area mainly in so far as they affect air quality inside livestock enterprises. The portion of small particles for long range transport (PM2.5) has been found to be in the range of 5% up to 70% from total dust also depending on the measuring procedure. To confirm this result, more particle size sensitive measurements will be done.

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Assessment of immission of dust and micro-organisms as part of permitting procedures for livestock management installations – legal basis and methods

Ewald Grimm [1] and Wilfried Eckhof [2]

Abstract

According to the German Federal Emission and Ambient Pollution Control Act (BImSchG, 2001) livestock installations shall be constructed and operated in such a way that this does not involve harmful effects on the environment and the neighbourhood. In addition, emissions must be reduced according to the state of the art. Potential environmental effects of livestock installations are assessed during permit procedures on the basis of the Technical Instructions on Air Pollution Control (TA Luft, 1986; TA Luft, 2002) or other regulations such as VDI guidelines. Usually the initial load of an air pollutant must be monitored or estimated and the additional load must be calculated by air dispersion modelling. The sum of both must not exceed defined immission limit values.

TA Luft (1986) is revised at the moment (TA Luft, 2002). As before, the regulation on emission limitation for total dust will not affect livestock installations. But in future a new limitation of PM_{10} immission load, which is based on the European Air Quality Daughter Directive (1999/30/EC) relating to limit values for NO_x, SO_2, Pb and PM_{10} in ambient air (European Union, 1999), will be in force. It is difficult to say to what extent animal farms will be affected by this regulation. According to it, immission of PM_{10} must usually be determined, if the emissions of a livestock installation exceed the so-called bagatelle-emission rate of 0,1 kg/h dust (equivalent to a livestock of about e.g. 900 fattening pigs and 8,300 layers) and if people permanently stay in the vicinity. Exceptions are possible if the level of the initial load can be estimated as “low” or if the level of the additional load caused by an installation is “irrelevant” in terms of TA Luft (2002) and additional measures for emission abatement are taken.

Up to now only few data on the PM_{10} immission load of rural regions where livestock production usually is located are available. At most rural sites in Germany it can be assumed, that the initial load is “low”. But there are exceptions, e.g. at sites in coastal areas (high level of natural sea spray) or at sites that are influenced by strong local sources or congested areas farther away. The additional load caused by an installation that is classified as irrelevant in terms of TA Luft (2002) is as low, that it is more or less a zero load. In those cases, where the initial load is not low and the additional load is not irrelevant an application however is possible, if it can be stated, that the sum of both does not exceed the immission limit values. For this, either data from public air monitoring networks are available and can be transferred or the initial load must be monitored at two sites over a period of at least one year. The additional load must then be calculated by air dispersion modelling.

Especially in permit procedures of intensive housing installations for poultry potential adverse health effects caused by the emission of dust, micro-organisms and endotoxins play an important role. The TA Luft (2002) only determines, that all possibilities should be assessed to reduce these emissions with measures according to the state of the art.

Referred to the assessment of environmental effects caused by micro-organisms, no generally confirmed methods or information are available. Nevertheless, authorities will have to decide upon. In practise, based on the results of numerous investigations, it is pragmatically concluded that at a distance of about 250 to 300 m from a livestock installation the immission load of micro-organisms is not increased compared to the background level. Usually it is sufficient to keep the setback distance for the prevention of odour nuisance according to TA Luft and/or VDI guidelines against dwelling houses in order to minimise health risks.

In order to confirm a specific setback distance regulation for minimisation of health risks, research is necessary on the casual relationship between emission, dispersion and immission and potential effects of dust and micro-organisms on the health of the immediate neighbours of livestock installations as well.

Keywords: emission, bioaerosols, animal husbandry, legal regulations, immission limit values

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1 Introduction

In the past, the emission and immission of dust did not play a role in licensing procedures of livestock installations in Germany for concentrations were far below the relevant threshold values for the protection of human health (e.g. TA Luft, 1986; VDI 2310 Bl. 19, 1992).

To improve air quality, the European Union has introduced the Air Quality Framework Directive (96/62/EC) on ambient air quality assessment and management (European Union, 1996). This Directive covers the revision of previously existing legislation and the introduction of new air quality standards for previously unregulated air pollutants such as particulate matter (PM_{10}). Based on the Framework Directive the first Daughter Directive (1999/30/EC) relating to limit values for NO_x, SO_2, Pb and PM_{10} in ambient air came into force in July 1999 (European Union, 1999). The Directive sets strict health limit values for PM_{10} for there is increasing evidence that negative health effects occur at very low levels of PM_{10} and without an apparent threshold (i.a. increasing rates of respiratory diseases and symptoms, of cardiovascular diseases and of mortality as well) (European Union, 1997). In this context type (e.g. organic, inorganic) and composition of dust is irrelevant. In Germany limit values for PM_{10} and other pollutants in ambient air are adopted not only in regulations on public air quality assessment but in regulations concerning the approval of industrial plants and other installations too.

Livestock buildings are a source of dust which mainly consists of PM_{10} (Seedorf and Hartung, 2002). Due to this, the limit values may be relevant for livestock installations too. In certain cases the amount of PM_{10} emitted, its immission and potential environmental effects would have to be determined and assessed as part of permit procedures in the future.

By comparison, the emission of micro-organisms (or so-called bioaerosols, which are a mixture of micro-organisms (e.g. bacteria, fungi) and metabolites (e.g. endotoxins) associated with dust) have already been a subject of discussion in permit procedures of intensive livestock installations in the past if the public was involved. Frequently objections are raised to the projects that emission of micro-organisms would cause health risks for the people living next to the farms.

This fears have been nourished by the so-called “Morbus Study” (Schlaud et al., 1998). Observed rates of respiratory diseases of children seemed to correlate to the intensity of livestock production. Because of the design of the study, a clear correlation could not be stated. There is still a lack of knowledge in this field.

Thereby emission of micro-organisms got subject of the considerations of the authorities and in the range of vision of politics. For the standard of knowledge is smattering (e.g. Seedorf and Hartung, 2002) the Association of German Engineers (VDI) has established several working groups. Their task is to document knowledge on measurement, emission, dispersion, immission and effects and abatement measures as well of microbiological air pollutants (Eikmann and Kämpfer, 1999). Priority is laid on the waste processing sector, for this is the predominating source of micro-organisms (Walter, 1999). In addition, a broad study on the assessment of health effects of bioaerosols from intensive livestock production is carried out in Lower Saxony (PROLAND, 2000).

In spite of the lack of knowledge and though there are no legally binding regulations such as limit values or assessment methods, potential environmental effects of the emission of micro-organisms have to be assessed as part of the permitting procedure of intensive livestock installations.

In this contribution the legal regulations and the way how the problem of emissions and immissions of PM and micro-organisms is dealt with in application procedures for livestock farms in Germany is described.

2 Legal framework with respect to the emissions to air in Germany

2.1 Overview

In Germany, the legal basis for the protection of human health and environment against aerial pollutants is laid down in the Federal Emission and Ambient Pollution Control Act (Bundes-Immissionsschutzgesetz – BImSchG, 2001). Corresponding requirements (e.g. emission and immission limit values, monitoring of pollutants, emission abatement techniques) are laid down in the First General Administrative Guideline Pertaining to the Federal Emission and Ambient Pollution Control Act - Technical Instructions on Air Pollution Control (Technische Anleitung zur Reinhaltung der Luft - TA Luft, 1986 and 2002). The requirements must be adhered to if industrial plants and other installations are constructed and operated, enlarged or in any other way substantial alterated.

In addition, guidelines established and published by the Association of German Engineers (VDI) play an important role in practise for the assessment of air quality and the reduction of emissions in livestock

farming (VDI 3471, 1986; VDI 3472, 1986; VDI 3474 (draft), 2001). The guidelines describe livestock farming techniques in general, the sources of emissions, feasibilities to reduce emissions and immissions and a method for odour assessment (minimum setback regulation) as well. For the guidelines focus on odour, they are not discussed in this paper.

The Maximum Immission Concentrations (MIK values) are determined by the VDI and published in VDI guideline 2310 (VDI 2310, 1974). Keeping within the MIK values ensures, according to current scientific knowledge and according to the appropriate criteria, that public health and the environment are safe. The values given in the guideline are guidelines, they have no force in law.

2.2 Main obligations according to the Federal Emission and Ambient Pollution Control Act (BImSchG)

According to the BImSchG (2001) livestock installations shall be constructed and operated in such a way, that this does not involve harmful effects on the environment or other hazards, considerable disadvantages and considerable nuisance to the general public and the neighbourhood (principle of protection). Referring to PM and bioaerosols, the neighbourhood must be safeguarded from harmful health effects.

In addition, precautions must be taken to prevent harmful effects on the environment, in particular by such emission control measures as are appropriate according to the state of the art (“Stand der Technik”, which is equivalent to “Best Available Technology”). According to the precautionary principle harmful emissions must be reduced by technical means below a certain limit. Limits depend on the hazardousness of a pollutant, the size of an installation, the technical feasibilities and the economic efficiency of abatement technologies.

2.3 Application of livestock installations

Construction, operation and expanding livestock and poultry production units as well as effecting substantial alterations regarding their operation are altogether subject to prior permits being issued by agencies legally responsible in such matters. During permitting procedure authorities will check whether a project complies with the obligations of BImSchG and TA Luft respectively.

Type and scope of permit procedures required depend on the kind and number of animals to be kept, on the specific unit's location and on possible or potential impact on the environment.

For animal housing proposed to be constructed, building permits must principally be applied for and obtained. In case animal production units reach or exceed sizes outlined in legal provision No. 7.1 of the regulation governing permission-based systems (4. BImSchV, 2001), a legal permit procedure according to the (German) BImSchG must be initiated and completed (table 1). A simplified application by the local authorities according to Building Law is sufficient for smaller units. In those cases public is not involved.

Column 4 of table 1 deals with units which, independent from type of stock and intended use, exceed 50 livestock units (1 LU is equivalent to 500 kg animal weight) and whose proportionate number per farmed area is above 2 LU per hectare. In this nexus, only such fields qualify as allowable which are regularly used by the farm in question.

At the beginning of permit procedures, the agency holding responsibility must first determine whether or not an assessment of a project's environmental impact (EIA) is necessary and hence warranted according to App. 1, Nos. 7.1-7.12 of the Federal Environmental Impact Assessment Act (UVPG, 2001; see table 1).

In the case of projects an EIA is not mandatory, the agency of cognizance first determines whether, based on the results of a preliminary examination, an EIA is needed. In this exercise, several criteria determined in the UVPG (2001) such as environmental pollution, nuisance potential and disturbance, ecological sensitivity of potentially affected areas, load-bearing tolerance/capacity of protected areas (e.g. nature conservation), extent of effects (geographic area/affected population) are given due consideration according to. Location-specific pre-examination means, that preliminary examination is to be solely performed on the basis of pertinent location criteria. By contrast, projects defined according to column 7 of table 1 do require due consideration of all criteria determined in the UVPG, in other words, which means that a so-called general preliminary examination is necessary. If after having reviewed a project the responsible agency comes to the conclusion that an EIA is in fact indispensable, a permission process involving the public must then be executed, even according to table 1 this would otherwise not have been necessary. In all other cases, simplified proceedings where the public is not involved, will do.

Table 1:
Permit-relevant project/agricultural unit capacities

Animal Species	4. BImSchV, App. No. 7.1			UVPG, App. 1, Nos. 7.1 - 7.12		
	Permission according to BImSchG			Environmental impact assessment (EIA)		
				Mandatory	Possible after individual case pre-examination	
	Public involved	Public not involved			Location-specific	General
Finishing pigs (≤ 30 kg)	2,000	1,500		2,000	1,500	
Sows (incl. piglets < 30 kg)	750	560		750	560	
Piglets (growing period, 10-30 kg)	6,000	4,500		6,000	4,500	
Layers	20,000	15,000	Unit size	42,000	15,000	Unit size
Pullets	40,000	30,000	> 50 LU	84,000	30,000	> 50 LU
Broilers	40,000	30,000	> 2 LU	84,000	30,000	> 2 LU
Turkeys	20,000	15,000	per ha	42,000	15,000	per ha
Cattle (> 2 years)	350	250		350	250	
Calves (< 1 year)	1,000	300		1,000	300	
Furred animals	1,000	750		1,000	750	

2.4 Course and content of permitting procedures

During permitting procedures authorities will check whether the farmer has met crucial obligations according to the BImSchG as concretised in the TA Luft. Additionally, establishment and operation must not conflict with any other provision under public law (e.g. nature conservation). If prerequisites are given, there is a legal obligation to grant the permission.

In the case of a permitting procedure according to the BImSchG, the building permission is included. Application forms include in particular general information on design and operation of an installation. A detailed description of the project (e.g. species and number of animals to be kept, housing and waste management systems) and information on emissions (e.g. type and quantity of emissions, location and dimension of sources) must be stated. Measures to reduce emissions and to avoid environmental effects must be specified. An assessment of environmental effects (usually on odour, but possibly on ammonia, noise and bioaerosols too) is carried out according to the regulations laid down in the Technical Instructions on Air Pollution Control (TA Luft) or other provisions.

Environmental impact assessment further affords a detailed description of the site (e.g. human beings, fauna and flora, soil and water) with respect to sensitivity and need of protection. The potential effects of the project on human beings and environment and interactions as well shall be identified, prognosticated and assessed. Finally, measures to avoid, reduce and compensate effects on the environment shall be described.

During the permitting procedure the enforcement authority will involve other authorities, e.g. for air pollution control and for nature conservation. Their statements get part of the permission. As described above, not only other authorities competence of which is affected, but also public must be informed and involved if serious effects on the environment are suspected. Documents must be open to the public. A meeting must be summoned to give public opportunity to discuss the project. Statements of authorities and public shall be taken into account when deciding upon approval.

3 Technical Instructions on Air Pollution Control (TA Luft)

3.1 General provisions

TA Luft (1986, 2002) serves for the implementation of the requirements of the BImSchG in permitting procedures of installations. It makes provisions for the protection of the neighbourhood and the environment against high loads caused by air pollutants. In this context not only the additional load caused by the own project, but the initial load caused by other sources such as industry or livestock installations must be taken into account. In addition, the chapter on emissions prescribes limit values for several pollutants and requirements for the construction and operation of installations in order to reduce emissions as well.

Table 2:
Concentration and mass flow rate of total dust from different types of livestock (Seedorf and Hartung, 2002; Takai et al., 1998)

Animal species	Concentration [mg/m³]		Mass flow rate	
	Median levels	Maximum levels	[mg/(animal h)]	of 0,2 kg/h is equivalent to .... animals
Pigs	2.08	6.2	111	1,800
Poultry	2.45	8.1	12	16,667
Cattle	0.66	1.5	110	1,818

The TA Luft (1986) which has come into force in 1986 is revised at the moment (TA Luft, 2002). Thereby the requirements for the protection of human health are expanded on the limitation of PM_{10} immission load, see table 3. By this way the European Air Quality Daughter Directive (1999/30/EC) relating to limit values for NO_x, SO_2, Pb and PM_{10} in ambient air is adopted to German law (European Union, 1999).

For the state of knowledge is not sufficient at the moment, there are no specific regulations for the limitation of the immissions of micro-organisms.

3.2 Limitation of emissions

Limit values of TA Luft (2002) apply to the emissions of total dust. Mass flow rate and concentration must not exceed 0,2 kg/h and 20 mg/m³ respectively. The concentration of dust inside and in the waste air of animal housings operated according to the state of the art usually is far below this limitation (table 2). Thus, livestock installations are not affected by it. Whereas, if only the mass flow rate would be limited, at least larger livestock installations (e.g. 1,800 pigs) would have to monitor and to reduce emissions. Nevertheless, stricter limit values have to be obeyed according to animal welfare and labour protection regulations (e.g. general limit value of 6 mg/m³ for fine particles (PM_5) relating the air in workspace (HVBG, 2001)). For particles are deposited in the stacks, the concentration inside buildings is higher than in the waste air (Platz et al., 1995).

Besides the limitation of total dust and in order to take the fears of the public against adverse health effects into account, all possibilities should be assessed to reduce emissions of micro-organisms and endotoxins with measures according to the state of the art (No. 5.4.7.1 TA Luft (2002)).

In principle, all measures to reduce dust emissions will lead to lower emissions of micro-organisms and endotoxins too, for dust is a carrier of micro-organisms. Measures include e.g. dry and clean housings, non-littered or scarcely littered housings, the use of dry and clean litter in the case of littered housings and cage-housing with aerated manure belts. But altogether viewed, the state of knowledge referred to abatement measures is insufficient yet.

In principle, gaseous and particulate emissions from animal housings can be reduced by waste air treatment (biofilters, bioscrubbers, chemical wet scrubbers or combinations of these technologies). For these devices cause high investment and operating cost so that an economic production is not possible, they are not state of the art of livestock farming in general. Therefore, TA Luft prescribes their application only secondary if prescribed setback distances are not met. In addition, these techniques can only be applied on fully channeled (mechanical) ventilation systems and not on naturally ventilated housings.

Reduction efficiency for micro-organisms and endotoxins depends very much on the quality and management of the filter and of the washing water, which is used to remove most of the dust from the air before entering the devices. If filter and washing water are contaminated, the microbial emissions from the filter can be several magnitudes higher for certain micro-organism species than in the animal house air (Hartung, 1999).

3.3 Immission of fine particles

The TA Luft (2002) determines limit values for the immission of fine particles (PM_{10}), which are cited from the European Air Quality Daughter Directive (1999/30/EC) (table 3). In contrast to the Directive, no margin of tolerance and no period to meet the limit values are admissible. Health limit values must be met instantly after TA Luft (2002) has come into force even far before 2005, which is the deadline according to the Directive.

In comparison, the immission limit values for total suspended particulate matter (TSP) are given in table 3. The immission concentration of PM_{10} is approximately 0.83 of the TSP-concentration (Hesse, 2000).

Table 3:
Immission limit values of TA Luft (2002) for PM_{10} and of TA Luft (1986) and VDI 2310 Bl. 19 (1992) (MIK-levels) for total suspended particulate matter (TSP)

Regulation	Concentration limit value [µg/m³]	Averaging period	Remarks	Level of initial immission load classified as "low" [µg/m³]	Level of additional immission load classified as "irrelevant" [µg/m³]
TA Luft (2002)	40	1 year	-	< 34	1.2
	50	24 hours	≤ 35 exceeding per year	50*	-
TA Luft (1986)	150	1 year	-	-	1,5
	300	24 hours	98th percentile	-	-
VDI 2310 Bl. 19 (1992)	500	1 hour	Up to 3 successive hours	-	-
	250	24 hours	-	-	-
	150	24 hours	On successive days	-	-
	75	1 year	-	-	-

*) over the last 3 years; ≤ 15 exceeding per year

TSP limit values according to TA Luft (1986) and VDI 2310 Bl. 19 (1992) are not relevant for livestock installations, for immission loads in the surroundings are usually far below these levels. Due to the immission limit value regulation for PM_{10} of the TA Luft (2002) and depending on the specific site conditions, monitoring of the initial immission load and prognosis of the additional immission load caused by an livestock installation applied for may be necessary in some cases in the future. The assessment follows the scheme illustrated in figure 1.

Immission of PM_{10} must usually be determined, if the emissions of a livestock installation exceed the so-called bagatelle-emission of 0,1 kg/h dust and if people permanently stay in the vicinity.

The bagatelle-emission is equivalent to a livestock of about e.g. 900 fattening pigs and 8,300 layers in conventional housing systems. It must be noticed, that livestock housings are treated as diffuse sources. This means, that the bagatelle-emission is only 10% compared to an industrial source with high stacks. In practise, "vicinity" is usually defined as a region within a radius of about 2 km from an intensive livestock installation.

Exceptions from the duty to determine the immissions are possible in order to simplify the execution of the regulation: First, if the level of the initial load can be estimated as "low" and second, if the level of the additional load caused by an installation is "irrelevant" (table 3) and additional measures for emission abatement are taken.

3.3.1 "Low" initial immission load

Preliminary assessment of the initial immission load of PM_{10} in ambient air should be based on the results of public ambient air monitoring networks according to the official arguments of the Federal Government. In addition, i.a. the results of former monitorings and of monitorings from similar areas could be used. Additional measurements by the applicant should only be ordered if there is reason to believe, that other sources such as quarrying or cement and lime manufacturing contribute to local loads. Monitoring is not necessary, when after evaluation of results from public air monitoring networks and prognosis or estimation of the additional load, it is evident that the immission limit values is adhered to.

To what extent the regulations will be relevant in execution is difficult to estimate. Only few data on the PM_{10} immission load of rural regions, where livestock production is usually located, are available. Most measuring sites are situated in congested urban areas and are influenced by heavy traffic.

In general, the load of PM_{10} reduced about 50% in the last 20 years. The actual level is about 25–50 µg/m³ (annual means) over extended area. The mean load is about 11-25 µg/m³ at sites located in the rural environment (UBA, 2000). This level complies with the data reported by the federal states of Germany, see table 4.

Highest levels are usually monitored in congested urban areas and at sites influenced by and close to major roads. But there are exceptions from this rule, as data from Lower Saxony indicate. For example, in coastal areas high loads could be caused by natural

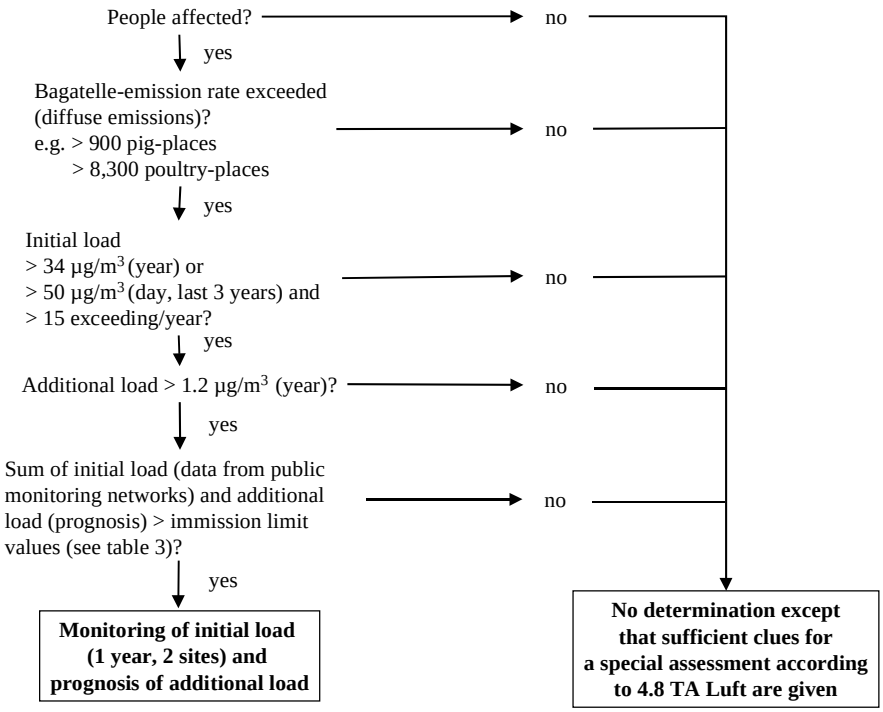

Figure 1:
Assessment scheme relating to PM_{10} immissions according to TA Luft (2002)

Table 4:
Concentration ranges of fine particles in ambient air at public monitoring sites in selected states of Germany

State (year)	Reported concentration ranges [µg/m³] (annual means)
Brandenburg (2001)	< 30 (rural sites < 20)
Lower Saxony (1999)	15 – 36 (urban sites 29 – 35, rural sites 17 – 29)
Northrhine-Westphalia (2000)	18 – 41 (congested urban areas)
	< 25 (remote sites)
Saxony-Anhalt (2000)	< 30 (only urban sites)
Saxony (1999)	14 – 34
Hesse (2000)	21 – 36 (10 – 17 remote sites)
Baden-Wuerttemberg (2000)	17 – 29 (urban sites; urban traffic sites up to 38)
	< 15 (background level)
Bavaria (2000)	21 – 38 (46 – urban site influenced by heavy traffic)

sea spray (Lower Saxony, 1999). Levels could also be influenced over larger distances by long range transport. A study carried out in Northrhine-Westphalia (Kuhlbusch et al., 2000) indicates in good agreement to the prevailing experience that urban areas are a bigger source of PM than rural areas. The study also reveals the influence that congested urban areas and busy motorways even in far distance (20-100 km) may have on the immission situation in rural areas by long range transport At most monitoring sites of the federal states the limit value for chronical immission loads is adhered to. In rural areas the immission concentration usually remains under the level of the TA Luft (2002) indicating a low initial load if there are no site specific conditions that have to be taken into account.

By comparison, the short-time immission load of PM_{10} is nation-wide the major problem. At many monitoring sites the daily mean limit value is more frequent exceeded as acceptable. In most cases this applies to urban traffic sites. But in rural regions too, exceeding may occur due to natural sources (e.g. Friesian and Friesian Islands; Lower Saxony, 1999), local sources or long range transport (Kuhlbusch et

al., 2000). At those sites daily means above 50 µg/m^3 may occur more frequent than 15 times a year so that the criteria of the TA Luft (2002) for a low initial immission load (table 3) is exceeded.

3.3.2 "Irrelevant" additional immission load

The additional load caused by an installation that is classified as irrelevant in terms of TA Luft (2002) is 1,2 µg/m^3. It is as low, that it is lost in the natural fluctuation of the background concentration, i.e. it is more or less a zero load.

To comply with this value, the mean concentration of dust in the air of e.g. a typical pig confinement building, which is about 2 mg/m^3 (Seedorf and Hartung, 2000), must be diluted by a factor of 1,000.

Up to now only few information is available about the emission amounts and the distribution characteristics of fine dust particles in the surroundings of animal houses that is based on measurements. Several studies conducted on different types of animal husbandry (up to 2,500 pigs, 15,000 hens and 20,000 chicken) revealed, that at a distance of approximately 115 m downwind of the housings the concentration of dust in the ambient air reached the level of the reference sampling points upwind (Schmidt and Hoy, 1996; Hartung et al., 1998). Platz et al. (1995) have measured fine particles (PM_5) and could not measure significant elevated immission concentrations at any point or time of the year in the surroundings of animal buildings. On the other hand Hartung et al. (1998) could prove by remote sensing (LIDAR), that emitted particles are transported even over a longer distance. However, many factors such as wind and weather conditions and the nature of the surrounding area (e.g. forest, meadow) have a considerable influence.

In addition, the type of ventilation system and the position of the air outlets have an influence on the transmission of the particles emitted. Compared to roof outlets deposition at close range to the building increases in case of housings with wall outlets and surrounded by an planted embankment (Krause, 1997).

The modelling of the dispersion of fine particles as prescribed in the TA Luft (2002) (Lagrange particle model) is based on the same assumptions as for ammonia concerning the velocities of deposition and sedimentation. In analogy to ammonia and in dependency on the topographical and meteorological conditions the distance, where the immission concentration reaches the level of the irrelevant additional load in the main wind direction, can be estimated up to 500 m in extreme cases. This finding is in contradiction to the results of the investigations cited above and needs further assessment.

4 Assessment of micro-organisms in practise

Micro-organisms and endotoxins associated with particles are aerial pollutants in farm animal housings that have been linked with several production diseases (infections of the respiratory system, allergic reactions in the airways of susceptible persons; Hartung, 1999). They are assumed to pose a risk for the health of farmers and workers in the farms and to the neighbouring residential areas around intensive livestock enterprises.

Potential adverse health effects caused by the emission of dust, micro-organisms and endotoxins are often on the agenda of permitting procedures of intensive housing installations especially for poultry. One reason may be, that concentrations and emissions of airborne micro-organisms are particularly high in poultry houses compared to cattle and pig houses (table 5 and 6).

In table 5 some data on typical concentrations of micro-organisms in the inside and outside air of agricultural sources compared to waste processing sources are summarised. In addition, typical background levels are given.

During permitting procedures, potential environmental effects caused by the emission of micro-organisms are usually assessed in a verbal-argumentative manner based on the actual state of knowledge. On the basis of the results of several studies the dispersion of micro-organisms at least can be estimated in tendency.

According to Hilliger (1984) the die-off rate of airborne micro-organisms in animal buildings is high, so that only about 80% of the micro-organisms that could be determined in the inside air get over into the waste air. Outdoors the micro-organisms underlie a multiplicity of atmospheric factors leading to rapid die-off (e.g. low humidity, high temperature, high UV-radiation, oxygen toxicity and pollutants). Only micro-organisms associated with particles have a chance to survive over a longer period of time. As described in context with the emission of fine particles, several factors such as weather conditions and the design of the ventilation system influence atmospheric transport of micro-organisms emitted.

Table 5:
Concentrations of micro-organisms in the inside air of different sources and in the ambient air in the surroundings

Type of source	Concentration [cfu/m^3]	Author
Different animal species' housings (without littering activity)	$1.3\ 10^5 – 9.1\ 10^5$	Rieger et al. (2001)
Cattle housings	$2.0\ 10^3 – 6.0\ 10^4$	Seedorf and Hartung (2002)
Pig housings	$9.0\ 10^3 – 8.0\ 10^5$	
Poultry housings	$2.0\ 10^3 – 1.5\ 10^6$	
Pig housings		Homes et al. (2000)
– inside	$3.0\ 10^4 – 2.9\ 10^5$	
– surroundings > 200 m	$3.0\ 10^2 – 8.0\ 10^2$	
Composting plants		Walter (1999)
– inside	$2.5\ 10^3 – 3.4\ 10^4$	
– surroundings (50 – 100 m)	$1.4\ 10^3 – 3.7\ 10^3$	
Pig / hen housings		
–inside	$9.0\ 10^3 – 8.5\ 10^4$	
–surroundings (50 – 100 m)	$4.0\ 10^2 – 1.2\ 10^3$	
Waste processing		Herr et al. (1999)
– Composting plants	$1\ 10^6$	
– Waste sorting plants	$1\ 10^5$	
– Dumping grounds	$1\ 10^5$	
– Refuse collection	$1\ 10^5$	
Surroundings of composting plants	$1\ 10^4 – 1\ 10^5$	
–up to 500 m	$1\ 10^2 – 1\ 10^3$	
–1,500 m		
Background level	$< 1\ 10^3$	Herr et al. (1999)
(urban and rural)	$1.2\ 10^2 – 3.0\ 10^2$	Walter (1999)

Table 6:
Mean emission rates for dust, micro-organisms and endotoxins (Seedorf et al., 1998; Takai et al., 1998)

Animal Species	Emission rates				
	Dust [mg/(LU h)]		Micro-Organisms	Endotoxins [µg/(LU h)]	
	Inhalable	Respirable	[log cfu/(LU h)]	Inhalable	Respirable
Cattle	145	24	-	-	-
- Cows	-	-	6.7	2.9	0.3
- Beef cattle	-	-	6.5	3.7	0.6
- Calves	-	-	7.4	21.4	2.7
Pigs	762	85	-	-	-
- Sows	-	-	7.6	37.4	3.7
- Piglets	-	-	7.2	66.6	8.9
- Fattening pigs	-	-	7.6	49.8	5.2
Poultry	3,165	504	-	-	-
- Layers	-	-	7.1	538.3	38.7
- Broilers	-	-	9.5	817.4	46.7

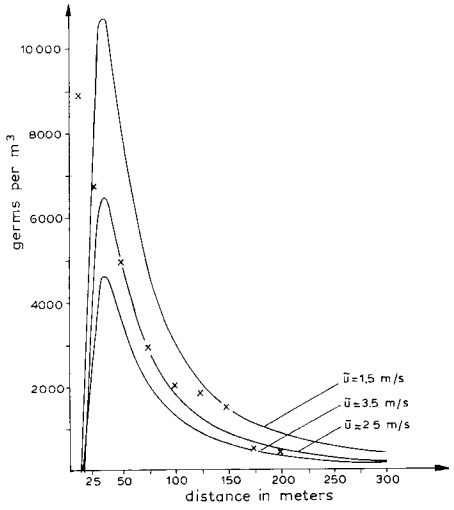

Figure 2:
Concentrations of bacteria in the surroundings of a hen house as calculated for 3 different wind speeds and measured (x) by Müller and Wieser (1987)

Müller and Wieser (1987) calculated a travel distance of viable bacteria from a laying hen house of 200 to 300 m downwind (figure 2). Similar results are reported by Hartung (1992). He investigated the concentrations of airborne micro-organisms and fungi during a whole year at 8 different sampling places in an area with high livestock density. He found higher concentrations at distances up to 150 and 250 m respectively from the source. Bacteria concentrations depended strongly on the distance and fungi concentrations were influenced by the season in particular. Hartung et al. (1998) confirm these foundings in general. Nevertheless, it is not precluded that particles and micro-organisms are transported farther. Measurements made around a farrow to finish swine production in the USA (500 sows) revealed, that at a distance up to 200 m downwind of the animal buildings increased concentrations of airborne micro-organisms occurred (Homes et al., 2000; table 5 and figure 3). Concentrations of total bacteria were consistently low 300 m from the buildings.

Other investigations carried out in the vicinity of piggeries and hen houses resulted no increased concentrations of micro-organisms in a range from 150 to 225 m (Hartmann, 1980). Hartmann states, that increased concentrations of micro-organisms are not to be expected if the setback distances according to TA Luft and VDI guidelines is adhered to and no excessive emissions emerge.

Platz et al. (1995) determined significant higher concentrations of stable specific micro-organisms up to a distance of 50 m downwind of different piggeries (360-2,500 fatteners) as compared to the background level upwind. They feel that a travel distance up to 100 m would be possible. To minimise the risk of infection by airborne bacteria the authors consider a minimum distance of 100 m and the setback distances according to the VDI guidelines respectively as reasonable.

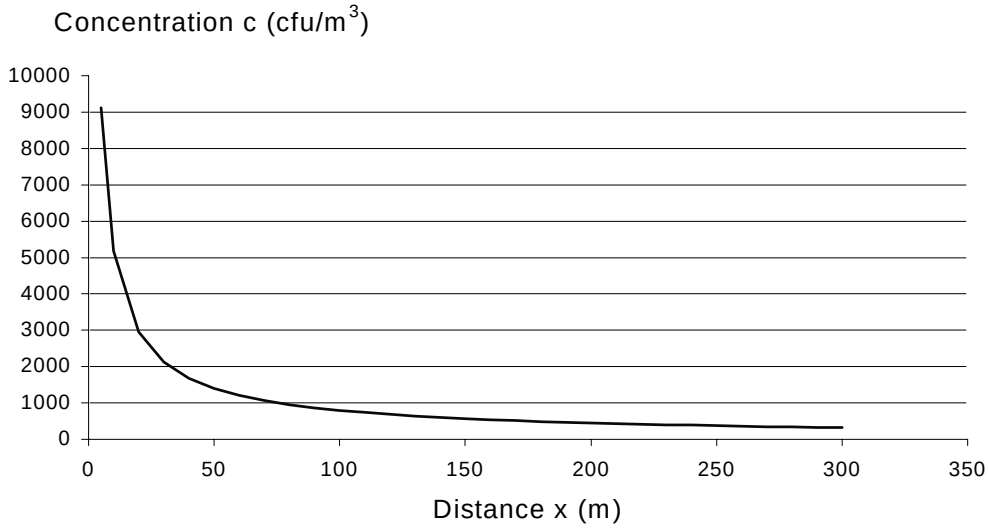

Figure 3:
Concentration of airborne micro-organisms in the surroundings of a combined swine production (curve fitting based on measurement data according to Homes et al., 2000)

In Austria comparative monthly measurements of the concentrations of micro-organisms were carried out at two livestock farms (600 pigs and 60,000 layers (cages)) and two composting plants (8,000 t per year

closed intensive composting and 1,200 t per year open composting) over a whole year (Walter, 1999). Compared to the background concentrations of micro-organisms at "unpolluted" rural and urban sites, she did not find increased levels in the vicinity of the animal farms. In contrast, the number of micro-organisms in the surroundings of the composting plants exceeded the background concentrations at a distance up to 150 m downwind. The authoress cites other studies according to which in dependency of the topographical and meteorological conditions the background level was reached at a distance of 300 m. Those studies revealed that the emission of mould is not generally a suitable assessment parameter for natural sources strongly influence the immission load.

5 Conclusions

The amended TA Luft (2002) prescribes stricter standards for the limitation and reduction of emissions and immissions of particles and their monitoring.

In practise the emission limit values of the TA Luft (2002) applying to total dust are not relevant for livestock installations for emissions are lower than these values. To reduce health risks for the neighbourhood emissions of micro-organisms and endotoxins should be reduced. But altogether viewed the state of knowledge on the abatement of the emissions of micro-organisms is insufficient yet and waste air treatment is not an appropriate measure up to now.

In contrast, the new regulations of the TA Luft (2002) concerning the limitation of immissions of PM_{10} may be relevant for the application of livestock husbandry. Although the immission load in rural areas generally can be expected as low, the regulation of the TA Luft (2002), which is thought to simplify the execution of the immission load regulation, probably does not take effect in all cases. The bagatelle-emission is as low that even installations larger than e.g. 900 pigs and 8,300 layers exceed it. In the case of alternative housing techniques such as littered pens or floor housing systems, that cause higher dust emission rates, the bagatelle-emission is equivalent to even a smaller livestock. In addition, the criteria of the TA Luft (2002) indicating a low initial immission load may be exceeded under specific site conditions (e.g. coastal areas with high natural sea spray concentrations, sites being influenced by congested urban areas or local sources). Finally, to assess whether the additional load is irrelevant or not, only few measurements have been carried out and results do not seem to comply with the results of air dispersion modelling. Further assessment is needed.

The adequate prognosis of the additional load caused by livestock installations with the help of air dispersion modelling is not possible for reliable and differentiated emission data are not available up to now. Measurements were often carried out under occupational safety and health aspects and corresponding sampling methods. Thus, a lot of data that have been published relate to the inhalable fraction (PM_{100} or total suspended dust) and/or the respirable fraction (PM_5). Hardly any data have been published for PM_{10}-emissions (e.g. Bayerisches Landesamt für Umweltschutz, 2000). No data are available for natural ventilated housings. In addition, it can be assumed that measurements based on separating techniques have usually not been carried out with an isokinetic sampling method for this requires an additional expenditure of labour.

If PM_{10} immission must be monitored and prognosticated according to TA Luft (2002) for otherwise it is not possible to prove that the immission levels are met, the initial load must usually be measured at two sites where immissions are supposed to be highest over a period of one year. The additional load must be calculated by air dispersion modelling. Cost resulting from this procedure can be estimated about 25,000 to 50,000 €.

Referring to microbial air pollution, investigations on the fate of micro-organisms emitted from animal houses revealed, that close to the buildings significant higher concentrations could be monitored. Several studies resulted that a distance of about 200 to 300 m from a livestock installation in tendency marks the border line, where compared to background levels, no increased concentrations occur. However, many factors such as weather conditions and the design of the ventilation system can have a considerable influence on the travel distance of micro-organisms.

The generalisation of the single findings on the dispersion of micro-organisms in the surroundings of livestock installations is restricted, for not all of the results of the studies are well-defined and the methods and conditions differ (esp. sampling methods, weather conditions). Nevertheless, tendencies could be identified that give indications for the practise in order to reduce health risks for residents. Assessment is usually proceeded on the assumption, that no increased levels occur, if the setback distances towards dwelling houses according to TA Luft or VDI guidelines (VDI 3471, VDI 3472 and/or VDI 3474) is adhered to (at least 250 to 300 m).

Nevertheless, there is still a considerable lack of knowledge on the amount of emissions from animal houses, on their dispersion in the ambient air, on immission and on potential health effects to the population living in the vicinity of animal enterprises, particularly in areas with high animal densities.

Finally it is worth to mention, that there is a conflict between the goals of animal welfare and environmental protection referring to the emission of PM and micro-organisms. As far as housing systems that are particularly animal friendly may lead to increased emissions compared to conventional systems (e.g. floor housing systems compared to cage housing systems, littered housings compared to liquid manure systems), they must keep a greater distance to the neighbourhood or the livestock must be reduced in order to meet immission limit values.

Altogether viewed, intensive research is necessary on the casual relationship between emission, dispersion and immission of dust and micro-organisms and on potential effects on the health of the immediate neighbours of livestock installations as well.

First of all, data on safe distances between farms and residential areas to prevent and to minimise risk of adverse health effects of dust and micro-organisms should be confirmed. There is equally a lack of knowledge on safe distances between farms in order to prevent transmission of infections that could cause animal diseases.

In particular, reliable and differentiated emission data of particles (esp. PM_{10}) and micro-organisms for different animal species, production sectors and housing systems (conventional, ecological) are needed. Sampling procedures for the measuring of emissions and immissions of airborne dust and micro-organisms must be standardised, in order to make results of different investigations comparable. This applies in particular to the measurements of emissions from diffuse sources such as natural ventilated buildings. Moreover action is needed to validate air dispersion modelling for micro-organisms (esp. die-off rate) and to develop measures for emission abatement of micro-organisms.

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Creating an inventory of agricultural PM emissions

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Abstract

Information from the literature, on agricultural activities and on emission factors, or failing those, on aerial concentrations, was used to compile a preliminary inventory of agricultural particulate matter (PM) emissions for the UK, with the aim of identifying optimum PM abatement strategies. The PM_{10} form of PM was chosen for the inventory, because of its relevance to the assessment of risks to human respiratory health. However, most of the data found in the literature were expressed in terms of inhalable particulates (broadly equivalent to PM_{tot}) and it was frequently necessary to use the very limited data available on the size distribution of different types of agricultural particulate to estimate PM_{10} emission rates or concentrations from PM_{tot} emission rates or concentrations.

The total gross agricultural emissions of PM_{10} from the UK in 1998 were estimated to be 17 kt (estimated lower bound 10 kt; estimated upper bound 24 kt). By far the largest contributor was housed livestock (12 kt). Of this, 9 kt were from housed poultry, 2 kt from housed pigs and 1 kt from housed cattle.

Unlike inventories of environmentally important gases, such as ammonia or methane, a PM_{10} inventory is mainly of use in protecting human health. It follows that the effect of 1 kg of emitted poultry PM may well be different from the effect of 1 kg of emitted soil PM, and so on. (By the same token, the *concentrations* of the emitted species may be more important, relative to emission rates, for PM than for gases.)

Ideally, a separate inventory should be compiled for each different type of agricultural PM (poultry, soil, grain and so on), but this would require many new measurements to be made. Also, much more work is needed to characterize the size distributions encountered with each different form of agricultural PM.

Keywords: Inventory, agriculture,particulate matter, PM_{10}

Introduction

Inventories of gaseous emissions (on a national, or other, basis) have been widely used now over a number of years, to identify the major sources of different gaseous pollutants, and hence to target research on abatement techniques. More recently, there has been interest (reference to TFEI) in extending this approach to emissions of particulate matter also. This paper reports and discusses work to produce a preliminary inventory of particulate matter (PM) emissions from UK agriculture.

It should be noted that whereas gaseous emissions are primarily of interest because they represent risks to the wider environment (e.g. acidic deposition in the case of ammonia, global warming in the case of methane and nitrous oxide), PM emissions are primarily of interest because they represent possible risks to the respiratory health of farm animals, of agricultural workers and of the public (Maynard and Howard, eds, 1999).

2 Methodology

The literature was searched for relevant information on agricultural activities (e.g. animal numbers, land areas) and for relevant emission factors for different agricultural operations. In the case of agricultural activities, much useful information could be drawn from existing inventories of emissions from UK agriculture of ammonia (Misselbrook et al., 2000), nitrous oxide (Chadwick et al., 1999) and methane (Sneath et al., 1997), as well as from the latest available UK agricultural census statistics (MAFF, 1999). Estimates were used where necessary: details are given, as appropriate, in Section 3, below.

The PM inventory itself was constructed using an Excel spreadsheet.

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3 Results

3.1 Summary of findings of literature review

3.1.1 Introduction

A large number of studies, in different countries, were identified, which report on some aspect of particulate emissions from agricultural activities. The review covered information from journals, from the United States Environmental Protection Agency (USEPA), and from communication with people working in the fields of animal and poultry husbandry and health, general farming practice, plant pathology, aerobiology, and compost production.

Quantitative estimates are given of emissions of PM from livestock buildings in Europe (Takai et al., 1998), and USEPA emission factors (deduced from large numbers of trials in the USA) for soil preparation, cereal harvesting, and unpaved roads. However, the majority of the other studies measured occupational exposure, and so are not necessarily representative of actual emissions. The various studies used different types of samplers, and results from different studies should be compared with caution. All measurements are subject to some degree of error, and even small changes in sampler position can cause a large difference to the results obtained, as can the environmental conditions such as humidity, windspeed, and temperature.

A further complication is that, although this study aimed to quantify particulate emissions in terms of PM_{10}, the majority of the studies did not measured amounts of PM_{10} itself. All studies measured mass of total suspended dust, and some measured respirable dust, which for most studies was taken as only the particles smaller than around 5 μm diameter. An estimate for PM_{10} and other size fractions can be made from this information (PM_{10} lies between total and respirable dust), by using an appropriate size distribution factor, but these factors are not well defined, and are different for different dusts.

The literature review outlined here is reported in much greater detail in the final project report (Project No WA 0802) submitted to MAFF, London (web address: www.defra.gov.uk).

3.1.2 Livestock buildings

A study by Takai et al. (1998) measured concentrations of inhalable and respirable particulate in the air of cattle, pig and poultry buildings in four European countries. Ventilation rates were also estimated for each building at each measurement, using a CO_2 balance method. This information was used to estimate emissions of particulates. Each building was measured over 24 hours, usually both in summer and winter, and was considered representative of current practice in each of the UK, Netherlands, Germany and Denmark. The results of these measurements are considered a good indication of current emissions from these buildings. However, measurements were not taken during operations such as moving animals or birds, or cleaning the buildings. These operations are relatively infrequent, but can generate high dust concentrations, albeit temporarily. The estimated emission from each type of building is shown in Table 1.

Estimates of particulate emissions from pig and poultry buildings were made in the Netherlands (Berdowski et al., 1997) which are somewhat higher than those reported by Takai et al. (1998). The Dutch estimates were made before the data from Takai et al. (1998) became available, and the Dutch authors state these are initial, first order estimates, to be taken as an indication of the scale of emissions, and that subsequent research suggests that actual emissions may be somewhat lower. A number of other studies have also looked at dust *concentrations* in animal buildings, in the UK, the USA, Finland, and Sweden, although these studies did not go on to estimate emission rates from their measured concentrations (not least because ventilation rates had not been measured). These studies were, rather, aiming to estimate exposure to workers, and so were not necessarily representative of *average* dust concentrations in buildings, as the animals are generally more active when farmworkers are in the buildings, and this increases instantaneous dust concentrations. No study other than that of Takai et al. (1998) measured particulate concentrations at night, which may partly explain why Takai et al. (1998) found consistently lower average concentrations of particulate. However, many factors influence the particulate concentrations within buildings, including: type of feed and feeding system, type of manure system, type and method of applying bedding (where used), temperature, humidity, activity of animals, and of course, ventilation rate. Wide variation between individual buildings has been found (*e.g.* Crook et al., 1991) so a large number of buildings would need to be measured in order to be confident that the sample is indeed representative of current practice in the UK.

The nature of dust from pig buildings in the USA has been thoroughly studied in two independent studies (Donham et al., 1986, and Heber et al., 1988). These found that the bulk of the dust was feed, but the finer material (particles smaller than around 10 μm) was mostly dried manure. Skin flakes, hair and other material were found in much smaller amounts.

Bedding was evidently not used in these pig units. Dust in poultry buildings is expected to be similar, with added contributions from litter (for meat birds) and feathers; however, further study of this would be beneficial. Particulate matter from cow and sheep buildings is rather different. Where good quality feed and bedding are used, particulate concentrations are low, and the majority of particles are fragments made from chopping and spreading the bedding. *In vitro* tests for different feed and bedding materials for horses found that hay is inherently dustier than alternatives such as silage, and straw is dustier than other bedding materials (Vandeput et al., 1997). If the bedding or hay is stored in a damp condition, very large numbers of moulds develop over time, and these come to far outnumber all other particulate material. This is particularly serious, because these moulds include species such as *Aspergillus fumigatus*, which are known to cause respiratory problems in farm workers and animals.

3.1.3 Arable farming

No published information, relevant to UK conditions, was found, from which a good estimate of particulate emissions could be made. However, the USEPA has calculated emission factors for cereal harvesting and soil preparation. The estimate for soil preparation requires the input of data concerning soil moisture and silt content; general average values for dry soil have been used, but, for more reliable estimates, UK data should be used.

Table 1:
Emission rates of particulates in kg per year per 500 kg liveweight of animal housed, from buildings in England (adapted from Takai et al., 1998)

Pig buildings 20/20*		Poultry buildings 12/12		Cattle buildings 16/4	
Inhalable dust	Respirable dust	Inhalable dust	Respirable dust	Inhalable dust	Respirable dust
5.5	0.82	27.5	3.3	0.85	0.27

* number of buildings surveyed in winter/summer.

Table 2:
Concentrations of particulates measured in air near agricultural operations

Reference	Country	Mean dust concentration, mg per m^3 of air			Comments
		Total suspended dust	Particles less than 10 µm diameter	Respirable particles (< 4 µm dia)	
<u>Soil preparation</u>					
Clausnitzer and Singer, 1996	USA, California			0.5 – 10	Depending on operation. Sampler <1 m above ground
Nieuwenhuijsen et al., 1998	USA, California	10-150	0.5 -9	0.1-1	Personal exposure with no protective cab, using cascade impactor for total and PM_{10}, and cyclone for respirable dust
Nieuwenhuijsen et al., 1999	USA, California	5		0.3	As above, using IOM sampler for inhalable dust
Norén, 1985	Sweden	100, 150			Mean dust concs at 2 points on outside of tractor
Batel, 1979	Germany	40			Mean concentration at tractor driver level. (Max conc was 630)
<u>Cereal harvesting</u>					
Clausnitzer and Singer, 1996	USA, California			1	Sampler 2.4 m above ground
Batel, 1979	Germany	20			Max conc 80. Sampler at tractor driver level
Darke et al., 1976	England		*2-10*		Approximate mass of spores collected behind combine

Several other studies have measured concentrations of particulates (or in one case, spores) near to arable operations. Studies performed in England, Sweden, Germany and Finland are considered to be representative of UK conditions: however results from California and the Canadian prairies are considered less relevant, because of significant climatic differences. A wide range of dust concentrations is reported, with several authors reporting values over 100 mg $m^{-3,}$ and mean concentrations well over 10 mg m^{-3} for some operations (see Table 2). Unfortunately, most of the studies are rather old, and were aiming to assess exposure to tractor drivers, who are nowadays enclosed in protective cabs. These reports do not all give complete details of procedure, numbers of measurements, or statistical treatments of results, so can only be interpreted as an indication of particulate concentration, rather than absolute values. Furthermore, great variations in measurements were found even with small differences in sampling location and weather. Thus, it is considered that emissions from arable farming could be significant. In order to perform a meaningful assessment, a large number of measurements, as part of a carefully prepared study, would be needed.

Emissions of biological particles, mainly fungal spores and pollen grains, can outnumber all other particles at certain times of year, particularly around harvest time, though their contribution to annual emissions is probably not high. There is not sufficient information to make a reliable quantitative estimate.

Wind erosion of soil is another source of particulate emissions which is probably very low on the national scale, but may be significant at certain times of year in local areas with soils prone to this type of erosion. Fine sandy and peaty soils are the types reported to suffer most from wind erosion: however, it is not known if PM_{10} emissions from these soils are significant, as the bulk of the particles are much larger. Research on this matter in the UK has been primarily concerned with reducing crop losses, rather than limiting emissions. This topic has been widely studied in the USA, but these data are not considered relevant to conditions in the UK, where the weather and soil conditions are very different.

The production of compost, whether for mushroom farming, or to turn raw organic wastes into more useful material, also has the potential to emit particulates, especially large numbers of spores of the microbes which are present in the composting process. The material remains moist at all times, and as long as it is undisturbed, emits only low levels. However, during turning, which is done regularly to aerate the material, a lot of steam and spores are released. Some studies on this subject concluded that this is certainly a potential occupational hazard to workers (who can become sensitised and develop allergic symptoms, though this is a fairly uncommon occurrence), but risks to the general public are not significant.

Various applications are made to arable land, to improve the quality or fertility of the soil, or to dispose of organic wastes such as manure or sewage sludge. The relatively small amount of slurry spread these days by gun spraying is obviously a potential source of emissions, though a study of the spread of pathogens through air was inconclusive. Poultry litter can be very powdery if dry, and applying this material to land could cause significant emissions to air. Lime is routinely applied, often in fine powder form, and is considered to be the application with the greatest potential for making significant emissions. Fertiliser application could also emit particulates, but most modern fertilisers are pelleted, so the potential for dust emissions is thought to be low. Pesticide application also has the potential to emit particles of either liquid or powder, particularly when applied to orchards, where the substance is entrained in air and blown onto the trees. Most other crops are sprayed by boom sprayers, which direct the substance down onto the crop and the chance for escape is small. The Code of Good Agricultural Practice for the Protection of Water (MAFF, 1998) specifies that pesticides ought not to be applied near watercourses in windy weather, to avoid contamination by the expected stray spray. Other applications are not expected to contribute significantly to airborne particulate emissions.

3.1.4 Post-harvest

Cereal drying is commonly carried out on the farm, and is reported to be a high emitter of particulates, though this is not covered by any regulations, and no reliable data have been collected. (Effective cereal drying is also necessary to limit the growth of moulds during storage.)

Transporting grain, milling, and producing animal feed are all processes which can potentially emit large quantities of particulates. The USEPA have developed emission factors for these, though it is not known how relevant they are to UK practices. In the UK, production of food and animal feed is covered by The Environmental Protection Act 1990, and emissions from plants are regulated by Local Authority Environmental Health enforcers. Some examples of emissions from some plants have been collected, but no UK nationwide data have been collated.

3.1.5 Combustion

Many farms have small incinerators used to dispose of items such as dead birds. Incinerators treating less than 50 kg/hour are not covered by UK regulations, but will nevertheless emit particulates. Data are not available. The UK ban on field burning of cereal straw has reduced emissions from arable farming, but some types of straw (e.g. linseed, which is difficult to bale) can still be burnt in the field. These will cause emissions of particulates which, although small on the national scale, may be significant locally.

3.1.6 Unpaved roads on farms

No UK data have been found for estimating of emissions from unpaved roads on farms, but this is certainly a potential source of emissions. The USEPA have calculated a formula for emission factor, and other studies in the USA found this was a reasonable approximation to measured values. Applying this formula to roads in a dry condition suggests that emissions from this source could be significant. Further study would be needed to check if this is definitely the case.

3.1.7 Energy use

Data are available to estimate PM_{10} emissions from energy and fuel use in agriculture, in 1998. The largest on-farm emissions come from burning straw as a fuel. Diesel and oil, used for vehicles, heaters and dryers, account for the next largest emissions of PM_{10}. Smaller emissions are generated by coal and gas. Use of electricity was estimated to produce significant amounts of PM_{10}, though this was produced at power stations, rather than at the farm.

3.1.8 Conclusions from the literature review

Most existing studies measured total suspended particulate or respirable dust, and interpolations to estimate PM_{10} can only be approximate. Also, estimates from all activities are as measured close to source, and a large proportion of this particulate is likely to be deposited nearby. Researchers at the University of California at Davis are starting to work to produce a factor to allow for this, and thus estimate net emissions from the farm. Further research on this topic is needed.

Livestock buildings appear to be the largest source of emissions of particulate matter from agriculture. Some reasonable estimations of these emissions are available, though further work would be valuable to improve on these estimates, and deduce more about the nature of these dusts in UK buildings.

Certain operations in arable farming, namely soil preparation and harvesting, can also emit large quantities of particulate matter. There are insufficient good data available to make realistic estimates of emissions, though a number of studies have measured dust concentrations, and report wide variations in results. Some emissions will also occur from applying material, in particular lime, to land, but no quantitative information is available. Other applications, such as manure, pesticide and fertiliser, may make a small contribution of particulate matter. At certain times of year, biological particles, such as pollen grains and fungal spores, can make significant contributions to particulate levels. Wind erosion of soils may be a significant source of particulates in some areas, but no suitable data have been identified.

Post harvest operations such as cereal drying, transport and handling, can also be high emitters of dust: some US emission factors are available, but not UK data. Preparation of animal feed is covered by IPC regulations. Emissions from unpaved roads can be predicted from a USEPA formula, which suggests that they may be significant, but verification in UK conditions would be needed. Energy and fuel use for agriculture has been well quantified, and emissions of PM_{10} are reasonably well known.

3.2 The preliminary inventory of PM_{10} emissions from UK agriculture

At the time of the study, the latest year from which UK agricultural census statistics (animal numbers, etc) were available was 1998. Thus the inventory reported refers to the year 1998.

It is stressed that the inventory described below is preliminary, as, except for housed livestock, the data on which it is based are very incomplete. There may, moreover, be other agricultural emission sources which are as yet unrecognised. There was also the problem that different authors have reported measurements on particulates in a wide range of different forms *e.g.* total dust, inhalable dust etc. To construct a meaningful inventory, it is of course necessary to "compare apples with apples and oranges with oranges", and a decision was therefore made to base the inventory on PM_{10} values, as a compromise between relevance to human health risks and availability of data. ($PM_{2.5}$ values are now believed to correlate better with human health risks than do PM_{10} values, but very few data on $PM_{2.5}$ were found.)

Where results were reported as PM_{10} itself, these were of course used for the inventory directly, but

where it was necessary to fall back on particulates reported in other forms, correction factors were used to convert these other forms to estimated PM_{10}. It is accepted that this is a crude procedure, but no better procedure was available. In the interests of transparency, the sources of all such correction factors, where used, were recorded.

Table 3 presents a summary of the resulting PM_{10} inventory, constructed using an Excel spreadsheet.

Table 3:
First estimate of total annual dust (PM_{10}) emission from UK agriculture (Year 1998)

Source	PM_{10} annual emission, t		
	Estimated lower bound	Average	Estimated upper bound
Housed livestock	6,900	11,500	16,100
Arable farming	800	2,400	4,000
Crop storage	10	20	30
Energy used on farms	2,300	2,900	3,500
Unpaved roads on farms	20	40	60
Total	10,000	16,900	23,700
% addition to total non-ag. PM_{10}	5%	8%	12%

As Table 3 shows, housed livestock contribute by far the largest proportion of total annual UK PM_{10} emissions. Of the total average of 11,500 t from housed livestock, 8,700 t come from housed poultry, 2,000 t from housed pigs and 800 t from housed cattle. The second largest term is from energy used on farms: here the largest contributor is the combustion of straw as a fuel in small boilers.

In the following paragraphs, comments will be made on the individual terms in the inventory and on our estimates of the upper and lower bounds on each component of the total emissions.

Thanks to the work of Louhelainen et al. (1987), a good value can be deduced, for animal house dust, for the ratio of PM_{10} to total dust (0.45) (and also of $PM_{2.5}$ to total dust (0.08)). Compared with some of the other sources of emission, the housed livestock emissions are fairly well characterized (Takai et al; 1998): an uncertainty band of ± 40% has thus been used. It is fortunate that this, the largest item in the inventory, has the smallest uncertainty bands.

For arable farming, no data reporting emission rates directly were found, but many data were available on dust *concentrations* arising near different arable operations. A simple model, as described below, was therefore used to estimate the former from the latter. The reported dust concentrations were assumed to prevail across the cross-section of a "plume" downwind of the dust-generating operation. A "plume" height of 3 m was assumed, and the "plume" was assumed to cover the whole UK field area for the operation concerned. For hay-making, four passes by a machine annually were assumed, but, for all other operations, only one pass annually was assumed. All the dust in each of these imaginary boxes was assumed to be emitted off the farm, with no re-deposition. For this source of emission, the plume height was believed to be the major source of uncertainty. Upper and lower bounds were therefore estimated by assuming extremes of plume height of 5 m and 1 m respectively.

In the case of crop storage, data reporting emission rates directly were found only for grain handling in the USA (Shaw et al., 1997). As with arable field operations, many data were found on *concentrations*, around other storage facilities but, unlike with arable field operations, no sensible estimate could be made of air flow rates around these facilities, so no attempt was made in this case to estimate emission rates from the concentration data. The uncertainty band on the grain handling term was estimated at ± 50%.

Energy use on UK farms includes off-road emissions from tractors and other vehicles, from heaters, dryers and other on-farm energy generation. The largest contribution comes from combustion of straw in small boilers. It has been assumed that all such boilers are in agriculture e.g for glasshouse heating, but this needs investigation. The statistics on straw burning boilers has not been revised since 1994, and the present numbers may be smaller.

Finally, for unpaved roads on UK farms, a simple model was used to estimate what total length of unpaved road might be found per unit of area of UK farmland, since the emission factors used, from the USEPA, are expressed as weights of dust per vehicle-

mile travelled. The reported total annual emission of 40 t PM_{10} assumes an average road surface moisture content of 10 %, which may be low by UK standards. If a value of 50 % is used in the USEPA formula instead, the total annual emission falls to 25 t PM_{10}.

4 Discussion

It must be noted that the PM_{10} emissions reported in the above inventory are (as with gaseous inventories) gross, not nett. In other words, no attempt has been made yet (because of lack of data!) to allow for re-deposition of emitted PM_{10}. It may well be that much of the emitted PM_{10} is re-deposited in the short range, e.g. within the farm on which it has been generated. Even the gross PM_{10} emissions from UK agriculture are estimated to be under 12% (see Table 3) of the total non-agricultural PM_{10} emissions, which are made up mainly of road transport and power station emissions.

There is a contrast, however, with gaseous inventories, in which, for example, emitted ammonia is the same, chemically, regardless of its specific source on the farm. For emitted PM, on the other hand, the *quality* may be very different from specific source to specific source. Thus PM_{10} from poultry may be very different chemically, and hence in its health effects, from PM_{10} from soil cultivation (Maynard and Howard, eds, 1999). Ideally, a separate inventory should be compiled for each different type of agricultural PM (poultry, soil, grain and so on), but this would require many new measurements to be made. Also, much more work is needed to characterize the size distributions encountered with each different form of agricultural PM.

Given that the health risks from emitted PM_{10} are probably better characterized in terms of the aerial *concentrations* they give rise to at different locations, there may in fact in the case of PM_{10} be merit in departing from the conventional concept of an inventory based on the *masses* emitted, and using instead “concentration maps”, covering the country or region in question.

5 Conclusions

A preliminary inventory of *gross* PM_{10} emissions from UK agriculture has been compiled. The total gross agricultural emissions of PM_{10} from the UK in 1998 were estimated to be 17 kt (estimated lower bound 10 kt; estimated upper bound 24 kt). By far the largest contributor was housed livestock (12 kt). Of this, 9 kt were from housed poultry, 2 kt from housed pigs and 1 kt from housed cattle. These, then, are the areas where research on abating PM_{10} emissions from agriculture would best be concentrated.

Unlike inventories of environmentally important gases, such as ammonia or methane, a PM_{10} inventory is mainly of use in protecting human health. It follows that the effect of 1 kg of emitted poultry PM may well be different from the effect of 1 kg of emitted soil PM, and so on. (By the same token, the *concentrations* of the emitted species may be more important, relative to emission rates, for PM than for gases.)

Ideally, a separate inventory should be compiled for each different type of agricultural PM (poultry, soil, grain and so on), but this would require many new measurements to be made. Also, much more work is needed to characterize the size distributions encountered with each different form of agricultural PM.

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European control strategy for fine particles: the potential role of agriculture

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Abstract

Robust associations were found between human exposure to fine particulate matter (PM) and a variety of health-related endpoints, such as cardiovascular diseases and premature mortality. Considering that fine particles can be transported with the air mass over long distances, the issue of controlling concentrations of fine particles was recently taken up at the international level by the European Union and United Nations Convention on Long-range Transboundary Air Pollution. Both bodies aim for international agreements on harmonized Europe-wide control strategies by 2004/2005.

This paper discusses efforts to develop a long-term European strategy to control fine particles. Preliminary estimates of present and future PM emissions in Europe are presented and the role of agriculture is outlined. Already now agriculture is an important, but not the only, contributor to European anthropogenic emissions of primary and secondary particulates. In absence of specific control measures taken in the agricultural sector, its relative contribution to total emissions will grow in the future, while stringent emission reductions will be implemented at other sources.

Major sources of PM from agriculture are poultry and pig housing, representing about 57 and 32 percent of PM10 emissions and 50 and 30 percent of PM2.5, respectively. According to the model estimates, the share of primary PM10 emissions from agriculture increased in the EU countries between 1990 and 2000 from 4.6 to 8.6 percent and it is expected to grow further to nearly 12 percent by 2010. Agricultural emissions of PM2.5 show similar, although slightly slower, growth.

Ammonia is an important precursor emission to the formation of secondary particles. Agriculture is the main source of ammonia emissions and attention needs to be paid to the possibilities to control sources of these emissions in the future.

Keywords: Modelling, particle emissions, primary and secondary particles, agriculture, European policy

1 Introduction

There is increasing recognition of the threat to human health posed by fine particulate matter. Robust associations were found between human exposure and particulate matter with aerodynamic diameters of less than 10 µm (PM10) and 2.5 µm (PM2.5) and a variety of health-related endpoints, such as cardiovascular diseases and premature mortality (Dockery *et al.*, 1993; Pope *et al.*, 2002). Fine particulate matter in ambient air originates from primary emissions of fine particles from various natural and anthropogenic sources as well as from the formation of secondary aerosols from precursor emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and volatile organic compounds (VOC). The typical residence time of the fine fraction of particulate matter ranges between 10 and 100 hours, during which such aerosols are transported with the air mass over long distances. Thus, as with other transboundary pollutants, fine particles at a given site originate from emission sources in a large region, typically including sources in other countries.

2 European Policies to Control Fine Particulates

Recognizing the strong transboundary component, the issue of controlling ambient concentrations of PM was taken up at the international level by the Commission of the European Union and the United Nations Economic Commission for Europe's Convention on Long-range Transboundary Air Pollution (UN-ECE/CLRTAP). Both bodies aim for international agreements on harmonized control strategies in 2004-2005. These agreements should be based, to the maximum possible extent, on substantive information about the effects of fine particulate matter to human health. To maximize the cost-effectiveness of the strategy, measures for reducing the various (primary and secondary) precursor emissions to PM, should be balanced across all contributing economic sectors including agriculture, and important side impacts on other environmental problems (e.g., acid deposition, ground-level ozone, etc.) should be taken into account.

The Convention on Long-range Transboundary Air Pollution has developed a detailed workplan to harmonize its various scientific activities to bring

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together scientific information about the effects of fine particles, the formation and dispersion of aerosols in the atmosphere, about the emission sources and their control potential. The synthesis and integration of this information should facilitate Parties to the Convention to include in 2005 the issue of fine particulate matter into the revision of the Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (UNECE, 1999).

The European Commission has launched the 'Clean Air for Europe' (CAFE) program to prepare a solid basis for the next revision of the air-quality related EU legislation in 2004/2005 (CEC, 2001). At that time, the Commission will review its various Daughter Directives of the Air Quality Framework Directive, revisit and, if found necessary, adjust the air quality limit values, update the Directive on Emission Ceilings and amend source-specific emission legislation. The Commission has stated that priority will be given, inter alia, to concerns about fine particulate matter. CAFE will systematically compile and review relevant scientific information to guide practical decision processes.

Both policy processes will apply the cost-effectiveness principle to determine the need for and the allocation of further emission reductions. Thereby, all required emission controls should be justified by achieving actual environmental improvements at least costs. This implies that emission controls should be economically balanced across all pollutants, emission sources and sectors that contribute to an environmental problem, so that overall costs are minimized.

As a practical tool, integrated assessment models will be used to analyze the cost-effectiveness of emission control strategies and to assist in the quantification of emission reduction requirements.

3 The RAINS Integrated Assessment Model

The Regional Air Pollution Information and Simulation (RAINS) model developed at the International Institute for Applied Systems Analysis (IIASA) is an integrated assesment model that brings together information on economic development, the structure, control potential and costs of emission sources, the formation and dispersion of pollutants in the atmosphere and an assessment of environmental impacts of pollution. RAINS addressses health-impacts and vegetation damage caused by ground-level ozone, acidification of terrestrial and aquatic ecosystems and eutrophication (excess nitrogen deposition) of soils. RAINS describes the inter-relations between these multiple effects and the range of pollutants (SO_2, NO_x, VOC and NH_3) that contribute to these effects at the European scale. A detailed description of the RAINS model can be found in Schöpp *et al.* (1999).

As discussed above, several emission sources contribute via various pathways to the concentrations of fine particulate matter in ambient air. While a certain fraction of fine particles found in the ambient air originates directly from the emissions of those substances (the "primary particles"), another part is formed through secondary processes in the atmosphere from precursor emissions, involving SO_2, NO_x, VOC and NH_3. Consequently, the search for cost-effective solutions to control the ambient levels of fine particles should balance emission controls over the sources of primary emissions as well as over the precursors of secondary aerosols. Thus, the control problem can be seen as an extension of the "multi-pollutant / multi-effect" concept (Table 1).

Table 1:
Air quality management as a multi-pollutant / multi-effect problem

	SO_2	NO_x	NH_3	VOC	Primary PM
Acidification	√	√	√		
Eutrophication		√	√		
Ground-level ozone		√		√	
Health damage due to fine particles	√	√	√	√	√
	Via secondary aerosols				

Estimating Emissions of Fine Particles

RAINS calculates emissions of particulate matter for three different size classes, i.e., smaller than 2.5 µm, particles between 2.5 and 10 µm, and larger than 10 µm. Thereby, PM10 is calculated as the sum of the first two classes and total suspended particles (TSP) as the sum of all three. The methodology includes three steps:

In a first step, country-, sector- and fuel-specific "raw gas" emission factors for TSP are derived. For solid fuels the mass balance approach is used where ash content and heat value of fuels and ash retention in boilers is considered. For liquid fuels, industrial processes, transport and agriculture TSP emission factors are taken from the literature.

In a second step, "raw gas" emission factors for each of the size fractions are estimated. This is done based on size fraction profiles reported in the literature for a variety of installations. They are typically given for PM10 and PM2.5 and are fuel- and installation (sector)-specific. The typical profiles are applied to the country-, fuel- and sector-specific "raw

gas" TSP emission rates (see first step) to derive the size-specific emission factors used in RAINS.

In a third step, actual PM emissions are calculated for the three size fractions. For a given country, PM emissions of a given size fraction are calculated by applying a general formula across every fuel and sector, taking into account the application rates of control technologies and size fraction specific emission removal efficiencies.

$$E_{i,y} = \sum_{j,k,m} E_{i,j,k,m,y} = \sum_{j,k,m} A_{i,j,k} ef_{i,j,k,y} (1 - eff_{m,y}) X_{i,j,k,m}$$

where:

i,j,k,m	country, sector, fuel, abatement technology;
Ei,y	emissions of PM in country i for size fraction y;
A	activity in a given sector, e.g. coal consumption in power plants;
ef	"raw gas" emission factor;
eff	reduction efficiency of the abatement option;
X	implementation rate of the abatement option.

Details can be found in Lükewille *et al.*(2001) and on the RAINS web site http://www.iiasa.ac.at/~rains.

Primary PM Emissions from Agriculture

Several agricultural activities contribute to the emissions of particulate matter, e.g., livestock housing, arable farming, managing crops, energy use, burning of agricultural waste and unpaved roads. Currently, the RAINS model allows estimating emissions from livestock housing, fertilizer application, energy use in agriculture (small stationary combustion and mobile sources) and to some extent from handling of crops. The following section is related to livestock farming, which is believed to be the largest source of fine particles from agriculture (ICC &SRI, 2000).

Most of the measurements of PM concentrations were performed on poultry and pig farms (e.g., Takai *et al.*, 1998; Louhelainen *et al.*, 1987; Donham *et al.*, 1986 and 1989), which are believed to be the major source of PM from animal housing (Berdowski *et al.*, 1997; ICC &SRI, 2000). Dairy and beef cattle are less important. The predominant sources include feed and faecal material and possibly bedding. Lower contributions originate from skin, hair, mould, pollen grains and insect parts. The ICC &SRI (2000) review indicates that the mass median diameter of dust collected in pig and poultry buildings is in the range between 11 and 17 µm. The proportion of PM5 in the total dust of pigs and poultry farms was estimated at about four to 16 percent (e.g., Heber *et al.*, 1988; Louhelainen *et al.*, 1987; Cravens *et al.*, 1981). The RAINS model assumes for all animal categories, similarly to the ICC &SRI (2000) study, size fraction distribution after Louhelainen *et al.* (1987), i.e., eight and 45 percent of inhalable PM for PM2.5 and PM10, respectively.

For agriculture, RAINS PM makes use of the databases of its ammonia module (Klimont, 1998), distinguishing between animals kept on solid and slurry systems (for cows, beef and pigs), laying hens, other poultry, and aggregating other animals into one category. Emission factors for animal housing derived from Takai *et al.* (1998) are supplemented with data about the length of the housing period (for cattle and pigs) in each country in order to develop country-specific emission rates per animal per year. Other factors, such as the type of feed, ventilation, varying animal weights, or climatic conditions, are not taken into account. Emission rates for fertilizer application are based on CEPMEIP (2001).

Historical activity data were taken from the United Nations Food and Agriculture Organization (FAO, 2001) and International Fertilizer Industry Association (IFA, 1998). The forecast up to 2010 is based on the scenarios prepared for the negotiations of the UN-ECE Gothenburg protocol and for the Emission Ceilings Directive of the European Community (Amann *et al.*, 1998 and 1999).

4 Results and Discussion

The RAINS model estimates emissions of primary particulates and precursors of secondary particles. The results of recent calculations are shown in Tables 2, 3 and 6. Further, a comparison with other studies is presented in Tables 4 and 5.

Primary Emissions of Fine Particles

Total European PM10 emissions are estimated for 1990 at about 7.9 million tons and are expected to decline by nearly 55 percent by 2010 (Table 2). Agriculture contributes about 200 thousand tons throughout the period with exception of the mid 1990's where the economic restructuring in the Central and Eastern European countries led to a decline in emissions of the non-EU countries. Agriculture in the EU countries contributes about 95 thousand tons of PM10 per year.

Total emissions of PM2.5 amounted in 1990 to 3.8 million tons and are expected to decline by 50 percent until 2010 (Table 3). Agriculture contributes about 40 thousand tons per year, i.e., in relative terms less than for PM10.

These estimates suggest for the EU-15 an increasing share of primary PM10 emissions originating from agriculture, increasing between 1990 and 2000 from 4.6 to 8.6 percent and expected to grow further to nearly 12 percent by 2010 (Table 2). A similar picture, although at a lower rate, appears for PM2.5, where the agricultural share grew from 1.7 percent in 1990 to 2.4 percent in 2000 and is expected to reach 3.5 percent by 2010 (Table 3).

Table 2:
Emissions of PM10 in Europe between 1990 and 2010 and the contribution of the agricultural sector (RAINS estimates)

	Year		
Region	1990	2000	2010
Emissions (Tg)			
EU – 15	2044	1135	804
Non – EU	5818	3143	2785
Total Europe	7862	4277	3589
Share of agriculture			
EU – 15	4.6 %	8.6 %	11.9 %
Non – EU	2.0 %	2.8 %	4.0 %
Total Europe	2.7 %	4.3 %	5.8 %

Table 3:
Emissions of PM2.5 in Europe between 1990 and 2010 and the contribution of the agricultural sector (RAINS estimates)

	Year		
Region	1990	2000	2010
Emissions (Tg)			
EU – 15	1098	774	518
Non – EU	2665	1494	1365
Total Europe	3764	2268	1883
Share of agriculture			
EU – 15	1.7 %	2.4 %	3.5 %
Non – EU	0.9 %	1.2 %	1.6 %
Total Europe	1.1 %	1.6 %	2.2 %

Agriculture is a less important source in non-EU countries, although also there the relative contribution to total PM emissions is expected to increase (Tables 2 and 3).

The increasing relative importance of agriculture for PM emissions is less a consequence of growing emissions in this sector, but is mainly caused by stringent emission controls in other sectors (e.g., stationary energy combustion, mobile sources). Thus, while in 1990 PM10 exhaust emissions from road vehicles were three to four times higher than emissions from agriculture, in the year 2010 both sectors will emit similar quantities. In the EU, agriculture will then emit more PM10 than the power plant sector. For PM2.5 from primary sources, agriculture is expected to remain a minor source, since most of agricultural PM emissions are larger than 2.5 µm.

Within the agricultural sector, the major source of PM is poultry and pig housing, responsible for about 57 and 32 percent of PM10 emissions and 50 and 30 percent of PM2.5, respectively. The share of these categories remains rather constant over years and across regions, i.e., varying by only 1 to 3 percent between EU and non-EU.

The discussed scenarios do not assume introduction of any control options specifically targeting emissions of PM from agriculture. On the basis of currently available information it was not possible to design model control technologies with well-defined efficiencies, investment and operating costs, and applicability rates for European countries. However, such techniques exist and are already applied to some extent (ICC &SRI, 2000). It would be preliminary to speak about the size of the reduction potential for fine particulate matter emissions from agriculture but according to ICC &SRI (2000) study there are a number of options with estimated reduction efficiency for PM between 30 and 50 percent (e.g., feed modification) that could be widely applied in animal housing. Once, more detailed information is available it will be possible to evaluate the cost-effectiveness of these measures in the "multi-pollutant / multi-effect" framework of the RAINS model.

Comparison with Other Studies

There is only a handful of studies that included agriculture in national or international fine particle emission inventories. Berdowski *et al.* (1997) estimated the contribution of agriculture to total EU emissions of PM10 and PM2.5 for 1990 at nearly 11 and 13 percent, respectively, indicating however that this might be on the high end. Indeed, a comparison between that study and more recent work of CEPMEIP (2001) and the RAINS estimates for 1995 seem to confirm that (Table 4).

Table 4:
Comparison of PM_{10} and $PM_{2.5}$ emissions from agriculture in the European Union (EU)

	RAINS [a)]	CEPMEIP, 2001[b)]	Berdowski *et al.*, 1997[c)]
PM_{10} (Tg)	93	140	306
Share	*7.0 %*	*6.9 %*	*10.7 %*
$PM_{2.5}$ (Tg)	17	31	140
Share	*1.9 %*	*2.8 %*	*12.6 %*

Estimates for different years: [a),b)] 1995; [c)] 1990

Significant changes occurred for estimates of PM2.5, where recent studies revised both absolute levels and relative shares downwards. Table 6 compares for the United Kingdom the estimates for animal housing conducted by the studies mentioned above and a UK inventory (ICC &SRI, 2000). Again, the older work of Berdowski *et al.* (1997) shows higher absolute levels of PM and larger shares than the national study (ICC &SRI, 2000), the recent international CEPMEIP inventory (CEPMEIP, 2001) and the RAINS model estimates. The difference between the latter three studies cannot be explained only with different base years (1995 and 1998, see comment to Table 5), especially since animal numbers have not changed substantially during these years. While ICC &SRI (2000) and RAINS derive their emission factors from Takai *et al.* (1998), CEPMEIP relies on Dutch emission rates that are especially different for poultry. This has an immediate effect on the total estimate, as poultry represents more than 50 percent of emissions from animal housing.

Table 5:
Comparison of PM_{10} and $PM_{2.5}$ emissions from agriculture (animal housing) in the United Kingdom

	RAINS [a)]	ICC&SRI, 2000[b)]	CEPMEIP 2001[c)]	Berdowski *et al*,1997[d)]
PM_{10} (Tg)	10.5	11.5	16.0	30.0
Share	*5.3 %*	-	*6.0 %*	*11.0 %*
$PM_{2.5}$ (Tg)	1.9	2.0	3.6	13.0
Share	*1.7 %*	-	*2.2 %*	*7.3 %*

Estimates for different years: [a),c)] 1995; [b)] 1998; [d)] 1990

As indicated earlier, animal housing is not the only source of PM emissions from agriculture. The ICC &SRI (2000) study for the United Kingdom includes arable farming, crop storage, energy used on farms, and unpaved roads on farms. Emissions from these sources amount to about 3.7 thousand tons of PM10 in 1998, i.e., nearly one third of emissions estimated for animal housing (Table 5). Other inventories include some of these sources in other sectors (e.g., agricultural energy use in residential combustion and transport), and it is virtually impossible to separate them as often the necessary statistical information is missing. According to ICC &SRI (2000), unpaved roads and crop storage contribute only very small amounts of PM10, while arable farming is with about 800 tons a more important source.

Emissions of Ammonia

Chemical analyses of PM2.5 samples at urban and rural stations throughout Europe reveal a dominating contribution from secondary aerosols, typically between 60 and 70 percent of total PM2.5 mass (e.g., Visser *et al.*, 2001; APEG, 1999). Ammonia emissions play a critical role in the formation of such secondary particles, and ammonium sulfate and ammonium nitrate constitute often more than 50 percent of the mass of secondary inorganic aerosols. Agricultural activities are the main sources of ammonia emissions, and influence through this second pathway PM concentrations in Europe in addition to their direct PM emissions. Total European NH_3 emissions are expected to decrease until 2010 by 17 percent compared to 1990, following the caps on national total ammonia emissions of the Gothenburg Protocol and the EU Directive on National Emission Ceilings. Although an improvement, this decline is significantly lower than committed reductions for other pollutants (e.g., -63 percent for SO_2 emissions), so that also for secondary aerosols the importance of the contribution from the agricultural sector is expected to grow (Table 6). There are technical measures to further reduce ammonia emissions from agriculture but they tend to be fairly expensive and of limited applicability. It is necessary to search for ways of improving the efficiency and applicability, as well look for new ways of controlling ammonia emissions.

Table 6:
Emissions of SO_2, NO_2, VOC and NH_3 in Europe between 1990 and 2010 (Amann *et al.*, 1999), in Tg

	Year	
Pollutant	1990	2010
Sulfur oxide	37938	13974
Nitrogen oxides	23396	13975
Volatile organic compounds	22640	13590
Ammonia	7558	6279

5 Conclusions

The Commission of the European Union and the United Nations Economic Commission for Europe's Convention on Long-range Transboundary Air Pollution aim for international agreements on harmonized control strategies in 2004-2005. These agreements should be based, to the maximum possible extent, on substantive information about the effects of fine particulate matter to human health. To maximize the cost-effectiveness of the strategy, measures for reducing primary and secondary precursor emissions to PM, should be balanced across all contributing economic sectors including agriculture, and important side impacts on other environmental problems (e.g., acid deposition, ground-level ozone, etc.) should be taken into account.

PM emissions from European agriculture are not expected to grow in the next decade. However, in absence of specific control measures taken in the agricultural sector its relative contribution to the total PM will further increase, especially for PM10. This is mainly a consequence of stringent emission controls being introduced in other sectors (e.g., stationary energy combustion, mobile sources). Thus, while in 1990 PM10 exhaust emissions from road vehicles were three to four times higher than emissions from agriculture, in the year 2010 both sectors will emit similar quantities. In the EU, agriculture will then emit more PM10 than the power plant sector. For PM2.5 from primary sources, agriculture is expected to remain a minor source.

Within the agricultural sector, the major source of PM is poultry and pig housing, responsible for about 57 and 32 percent of PM10 emissions and 50 and 30 percent of PM2.5, respectively.

Agricultural activities are the main sources of ammonia emissions, which play an important role in the formation of secondary particles, and influence PM concentrations in Europe in addition to their direct PM emissions. Although, total European ammonia emissions are expected to decrease until 2010 by 17 percent, compared to 1990, this decline is significantly lower than committed reductions for other pollutants, so that also for secondary aerosols the importance of the contribution from the agricultural sector is expected to grow.

There exists a potential for controlling PM emissions from agriculture. This, however, was not considered in the presented calculations since only insufficient data on control options were available. Once, more detailed information is available it will be possible to evaluate the cost-effectiveness of these measures in the "multi-pollutant / multi-effect" framework of the RAINS model.

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Lagrangian dispersion modelling

Lutz Janicke [1]

Abstract

Lagrangian dispersion modelling is a technique in the field of numerical modelling of atmospheric diffusion. It is not based on the advection diffusion equation but simulates the trajectories of a sample of particles. From these trajectories the values of concentration and deposition are derived. It contains no calibration parameters but relies solely on meteorological parameters that can be determined without dispersion experiments. Nevertheless it shows excellent agreement with dispersion experiments.

Lagrangian dispersion modelling has been standardised in Germany by several guidelines. The new draft of the German TA Luft (Technical Instruction on Air Quality Control) that governs the licensing of facilities requests a model of this type for performing dispersion calculations.

Keywords: Lagrange, random walk, dispersion model, TA Luft

1 Introduction

Up to now, the most popular numerical models for calculating atmospheric diffusion of air pollutants or some tracer material are the Gaussian plume models. Here, the plume resulting from a pointlike source is modelled for a stationary situation according to experimental findings. One can think of this model as simply an interpolation scheme for experimental results. This model is very simple and inexpensive with respect to computer time. However, it is usually applicable to simple situations only and to parameter ranges, where experimental data are available.

Eulerian models are much more general. Here, a 3-dimensional computational grid is used and for each grid cell the mass balance of incoming and outgoing fluxes of the tracer considered are calculated using the advection diffusion equation. Reliable numerical schemes for solving this equation are rather complicated. In addition, certain important physical aspects of atmospheric diffusion are neglected by this model as we will see later on.

Therefore, the idea arose to simulate the trajectories of tracer particles immediately (Lagrangian picture) instead of investigating the fluxes (Eulerian picture). This approach offers much more flexibility and precision in modelling the physical processes involved and will be described in the following.

2 The concept of the model

Out of the huge number of particles (dust, aerosol, gas) usually emitted by a source, only a small sample is considered. The sample size N is typically of the order of 10^4 to 10^8 particles, depending on the problem and available computer resources. The i-th particle is emitted at some time t_i and follows the trajectory $\mathbf{r}_i(t)$. These trajectories are calculated on the computer.

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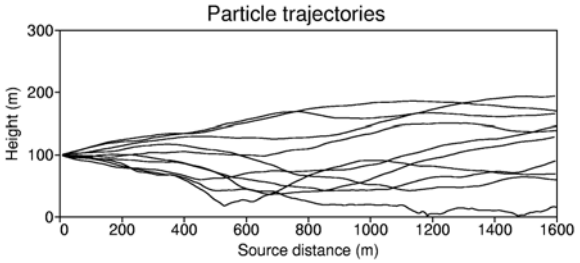

Figure 1:
Trajectories of 10 simulation particles emitted from a point like source at the height 100 m.

Each particle in the sample represents the mass m_i of a large number of particles and the sum over all these masses, Σm_i, is the total mass emitted by the source[2]. Usually the mass $m_i(t)$ changes with time due to removal or transformation processes.

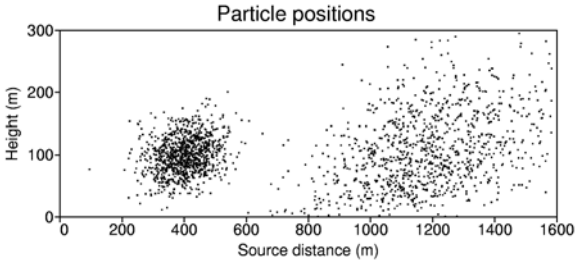

Figure 2:
A cloud of 1000 simulation particles emitted all at the same time at the height 100 m, shown 80 seconds after emission (left part the picture) and 240 seconds after emission (right part).

The mass concentration c at some time t and position **r** is calculated by a summation over the masses of all the sample particles that happen to be at time *t* in a small volume V around the point **r** and by dividing the sum by the size V of this volume,

$$c(\mathbf{r},t) = \frac{1}{V} \sum_{i|r_i(t)\in V} m_i(t) \qquad (1)$$

Hence, the mass concentration calculated represents a spatial average. In practice, also an averaging with respect to time is performed by means of counting all particles that visit the test volume V within a given time span Δt (a few minutes up to some days or years).

It is essential to allow for this averaging in order to reduce the sampling error. Otherwise the sample size has to be increased at the cost of an increased computation time required to run the simulation.

2 Instead of the mass m some other property of the matter emitted may be considered, e.g. radioactivity, odor or germ number

3 Details of the algorithm

The path of the *i*-th particle in the sample, the trajectory $\mathbf{r}_i(t)$, is determined by

- start time t_i and start position $r_i(t_i)$, depending on the configuration of the source,
- average wind field, depending e.g. on the profile of the terrain and the geometry of obstacles,
- atmospheric turbulence, depending e.g. on the atmospheric stability and the surface roughness,
- external forces (e.g. gravitational settling).

The mass $m_i(t)$ represented by the i-th particle in the sample depends on

- the mass emitted by the source(s) (might be time dependent),
- transformation and removal processes (e.g. dry and wet deposition, interception).

Each of these topics is handled by an appropriate sub-model, e.g. the initial parameters by an emission model and the removal processes by a deposition model. The most difficult part is the incorporation of atmospheric turbulence into the model. Several methods are conceivable:

A The turbulent wind field or the most important part of the turbulence spectrum is dynamically simulated (large eddy simulation). This is very time consuming and may be done only for a few special cases (e.g. Mason, 1994).

B The appearence of a turbulent wind field is simulated (kinematic simulation particle model, KSP). This is much cheaper with respect to computation time but yet in the research state (Fung et al., 1992; Yamartino et al., 1995).

C The action of the turbulent wind field on a particle is simulated by a random process (random walk model). There are fast algorithms available for performing random walk processes on a computer and a lot of research has been done on this type of model within the last 20 years (Wilson, 1982; Legg and Raupach, 1982; Janicke, 1983; van Dop et al., 1985, Sawford, 1986; Thompson, 1987; Janicke, 2000).

Therefore, Lagrangian dispersion models usually use a random walk process and trajectories generated in this way are shown in figure 1. For each particle, the position **r** and a random velocity **v** are calculated in a sequence of time steps of size τ:

$$r(t+\tau) = r(t) + \tau\left[\overline{V}(r) + \upsilon(t)\right] \quad (2)$$

$$\upsilon(t+\tau) = A(r,\tau)\cdot\upsilon(t) + B(r,\tau) \quad (3)$$

Here, $\overline{V}(r)$ is the mean wind field, $\mathbf{A}(\mathbf{r},\tau)$ is a function determined by the atmospheric turbulence and the time step, and $\mathbf{B}(\mathbf{r},\tau)$ is a velocity randomly chosen with each time step. Each particle is moved independently of the other particles. Therefore, correlation effects between particles and concentration fluctuations can not be modelled with this scheme.[3]

The physical quantities necessary to determine these parameters are (simplified):

$\overline{V}(\mathbf{r})$	mean wind field
$\sigma_{u,v,w}(\mathbf{r})$	turbulent wind fluctations
$K(\mathbf{r})$	diffusion coefficient

They are available from meteorological boundary layer models or from direct measurements. In any case, there are no calibration parameters in this model that have to be determined by dispersion experiments.

One important point in atmospheric diffusion is the fact that the random movement of a particle in a turbulent flow field is rather smooth, in contrast to the Brownian motion. A particle is moved by a turbulent eddy of typical size for some time (the Lagrangian correlation time T_L), before it is caught by another eddy. This is reflected by Equation (3), where the simulation particle preserves its turbulent velocity to some part with each time step. The parameter **A** determines how long the particle remembers its velocity and it is therefore strongly related to the Lagrangian correlation time T_L. The value of T_L varies between some seconds and a couple of minutes, depending e.g. on the velocity component considered, the height above ground, and atmospheric stability.

Therefore, the width σ_y of a plume generated by a stationary point source is proportional to the source distance x for travel times short with respect to T_L, as can be seen from figure 1, whereas classical diffusion would predict $\sigma_y \sim\sqrt{x}$. Atmospheric diffusion can be described by the classical advection diffusion equation for large distances only and not in the vicinity of the source.

4 Validation

Model validation is the assessment of how well the model performs. Typically, model predictions are compared to experimental results and the differences are evaluated. For atmospheric dispersion models this procedure is difficult because experimental results exhibit a large scatter due to the chaotic nature of turbulence. In addition, it is not possible to repeat an experiment exactly because the atmospheric conditions will always have changed to some part.

Therefore, from the many comparisons that have been performed, only a comparison with a wind tunnel experiment is shown here.[4] Experiments in the wind tunnel are usually restricted to neutral stratification, but they can be evaluated precisely and repeated exactly.

The comparison with the results calculated by the Lagrangian dispersion model AUSTAL2000 in figure 3 shows very good agreement.[5] Because of the free scaling of wind tunnel results, the situation modelled can be interpreted e.g. as the plume generated by a stack with a height H = 100 m in flat terrain with roughness length z_0 = 0.7 m (industrial site) or with H = 10 m and z_0 = 0.07 m (agricultural site).

When modelling the dispersion of coarse grained dust it may be necessary to modify the diffusion coefficient used. Gravitational settling of heavy particles shortens the time these particles spend within an eddy. Therefore, the Lagrangian correlation time decreases and so does the effective diffusion coefficient. This should become noticable when the settling velocity v_s is comparable to the vertical velocity fluctuations σ_w of the turbulence. Usually, this effect is neglected in dispersion models.

3 Concentration fluctuations can be modelled if a kinematic simulation of turbulence (type B in the listing) is applied

4 The data have been provided by the Meteorological Institute of the University Hamburg during the project Validation of the 'Kinematic Simulation Particle Model (KSP)' for Applications in the Context of German Air Quality Protection Laws, UFOPLAN-Ref. No. 98-295 43 354, funded by the Federal Environmental Agency, Berlin (1998)

5 Some ambiguity is introduced by the fact that the Coriolis force is not effective in the wind tunnel. This has been taken into account in the calculation presented

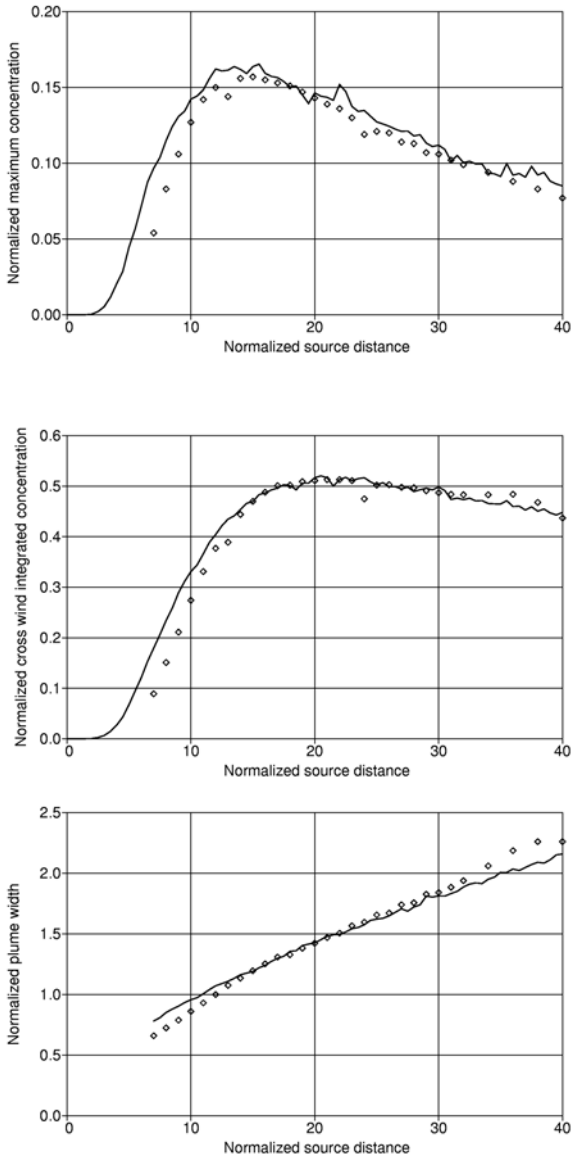

Figure 3:
Comparison between results from wind tunnel experiments (diamonds) and calculations with the Lagrangian dispersion model AUSTAL2000 (solid line). The ground level concentration field c(x,y) is analysed for the following situation: stationary point source (stack) of height H at x = 0 and y = 0 with source strength Q, flat terrain with roughness length z_0 = 0.007 H, neutral atmospheric stratification, wind velocity U at height H. The upper part shows the maximum normalized concentration $UH^2Q^{-1}c(x,0)$ as a function of the normalized source distance x/H. The part in the middle shows the normalized crosswind integrated concentration $UHQ^{-1} \int c(x,y)dy$ and the lower part the normalized horizontal plume width $\sigma_y(x)/H$.

5 Availability of the model

About 50 years ago in the field of hydrodynamics, the idea was formulated to model advection in numerical schemes by following the trajectories of simulation particles. Then, particle simulation had been widely used in plasma physics and astrophysics. About 20 years ago, a lot of research has been done on Lagrangian dispersion modelling in atmospheric physics and since more than ten years this technique has been used by experts in air quality control.

Nowadays, it is becoming common practice in this field. The first step was the formulation of a model standard by the German Association of Engineers (VDI) in the guideline VDI 3945 part 3 (2000). The meteorological profiles to be used in this model are given by the guideline VDI 3783 part 8 (draft, December 2001). This is the basis for the dispersion model formulated in the new German TA Luft (Technical Instruction on Air Quality Control, draft 2001-12-12) that governs the licensing of facilities. The Federal Environmental Agency provides the computer programlinebreak AUSTAL2000 for dispersion calculations complying with this rule.[6] The calculations presented in figure 3 were carried out using this program.

Some of the main features of AUSTAL2000 are:

- Use of meteorological time series
- Use of meteorological statistics
- Calculation of concentration and dry deposition (annual averages, highest daily or hourly values, time series)
- Gravitational settling of coarse dust
- 3-dimensional wind fields in complex terrain
- Point, line, area, and volume sources
- Arbitrary number of sources with time dependent source strengths
- Automatic estimation of the sampling error

A free computer program (with source code) is also provided by the VDI as a supplement to the guide line VDI 3945 part 3 (see http://www.vdi.de/partikelmodell).

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6 The program is distributed free of charge, see http://www.uba.de. Source code is available, the user guide is in german only

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Dispersion of germs from animal houses and their relevance to immission

Karl-Heinz Krause and Stefan Linke [1]

Abstract

The investigation of environmental load by emission from agriculture needs suitable dispersion models. Indeed there is no agro-physics, but there are special features in dispersion events of agricultural production that have to be taken into account. They start with wind induced sources and deflection conditions influenced by obstacles and end by unknown survival rates of germs and therewith vague risk assessment on immission site. Of course, for making decisions one cannot wait until all problems are solved irrevocably. But the pressure of administration to act may not lead to one-sided orientation as it happens at present in the model establishment of the TA Luft. We do not need imposed models but such that satisfy the reality conditions. Great deficits are observed with respect to agriculture.

Keywords: animal production, dispersion modeling, numerical simulation, risk assessment, TA Luft

Zusammenfassung

Zur Ermittlung der Umweltbelastung durch Emissionen aus der Landwirtschaft bedarf es geeigneter Ausbreitungsmodelle. Es gibt in der Tat keine Agro-Physik, wohl aber gibt es Besonderheiten bei den Ausbreitungsvorgängen in der landwirtschaftlichen Produktion, die es zu beachten gilt. Das beginnt bei wind- induzierten Quellen und hindernisbeeinflussten Ableitbedingungen und endet bei den unbekannten Überlebensraten von Keimen und damit einer vagen Risikoeinschätzung hinsichtlich ihrer Wirkung auf der Immissionsseite. Man kann bei Entscheidungsfindungen sicherlich nicht warten, bis alle Probleme unwiderruflich geklärt sind. Doch der Handlungsdruck in der Verwaltung darf nicht dazu führen, sich einseitig zu orientieren, wie derzeit bei der Modellfestschreibung in der TA Luft. Wir benötigen keine aufoktroyierten Modelle, sondern solche, die den Realitätsanforderungen genügen. Hier bestehen im Hinblick auf die Landwirtschaft sehr große Defizite.

1 Introduction

The agricultural production of plants and animals is characterized by emission of different air transported substances like gases, particles, germs, aerosols and several types of mixtures of them. While the emission rate in plant production is a seasonal one we have to recognize the nearly continuous output from animal houses. Both systems have in common that the airborne release takes place near to the ground. This means that structures of houses, plants and surfaces influence the transport mechanism of the emissions by the atmospheric wind. Furthermore the buoyant forces over the great areas of roofs of animal houses and over fields with different plant surface temperature cause vertical and horizontal motion at a small scale. Nevertheless, answers are expected to the questions where the emissions remain after they are released into the environment and what is their relevance to immission.

If the transport of airborne substances by the "vehicle" air is considered as mixture and not separately for multiphase flow, such a diffusion model is expressed by three mixture conservation equations of mass, momentum and energy and an additional one for concentration changes: the diffusion equation. Otherwise we have three field equations for each phase with three coupling jump conditions. Therefore correctly defined mixture quantities are of importance.

The mixture density ρ_M in a two-phase flow is given by

$$r_M = a\, r + b\, c \qquad (1)$$

c stands for the density or concentration of a single pollutant (phase 2), ρ is the density of the transport medium air (phase 1). The axiom of continuity requires

$$a + b = 1 \qquad (2)$$

With the definition of ρ_M and the mixture velocity vector $\mathbf{v}_M$

$$\mathbf{v}_M = \frac{1}{\rho_M}(\alpha\, \rho\, v + \beta\, c\, \mathbf{v}_C) \qquad (3)$$

1 Dr. Karl-Heinz Krause belongs to the scientific and Stefan Linke to the technical staff of the Federal Agricultural Research Centre (FAL), Institute of Technology and Biosystems Engineering (Directors: Prof. Dr. Axel Munack and Prof. Dr. Klaus-Dieter Vorlop) in the Federal Republic of Germany, Bundesallee 50, 38116 Braunschweig.

- $\mathbf{v}$ and $\mathbf{v}_C$ are the different phase velocities - the mixture continuity equation

$$\frac{\partial \rho_M}{\partial t} + \text{div}\,(\rho_M\ \mathbf{v}_M) = \alpha\left[\frac{\partial \rho}{\partial t} + \text{div}\,(\rho\mathbf{v})\right] + \beta\left[\frac{\partial c}{\partial t} + \text{div}\,(c\mathbf{v}_C)\right] = \tag{4}$$

leads with

$$\beta = O\,(10^{-5}) \tag{5}$$

to the well known continuity equation of incompressible flows with $\mathbf{v_M} = \mathbf{v}$

$$\text{div}\ \mathbf{v} = 0 \tag{6}$$

The diffusion equation (continuity equation) for phase 2 is expressed by a source term S of mass generation and a diffusion term with the diffusion velocity $\mathbf{v_{CM}} = \mathbf{v_C} - \mathbf{v}$

$$\frac{\partial c}{\partial t} + \text{div}\,(c\mathbf{v}) = \frac{S}{\beta} - \text{div}\,(c\mathbf{v}_{CM}) \tag{7}$$

It is a question of pragmatism how to formulate the mass fluxes of each phase. In the case of $\mathbf{v} = \mathbf{0}$ molecular diffusion dominates the dispersion behaviour.

$$\frac{\partial c}{\partial t} + \text{div}\,(c\mathbf{v_C}) = \frac{s}{\beta} \tag{8}$$

According to Fick's law the mass flux $c\mathbf{v_{CM}}$ is proportional to the gradient of the concentration of matter:

$$c\mathbf{v}_C = -\,D\ \text{grad}\ c \tag{9}$$

The coefficient of proportionality D is the (constant) diffusivity. The diffusion equation in Cartesian coordinates is

$$\frac{\partial c}{\partial t} - D\ \text{div}\,(\text{grad}\ c) = \frac{S}{\beta} \tag{10}$$

The operation div grad refers to the Laplace-Operator:

$$\text{div grad} = \Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \tag{11}$$

The molecular diffusivity D is of the order of 0.2 cm^2/s for air and 10^{-5} cm^2/s for water. Diffusion can be defined as an irreversible process by which matter, particles, germs, populations, etc. are distributed according to individual random motion. Each individual moves a short distance λ in an arbitrary direction in a short time τ. Diffusion occurs from high concentration to low concentration. λ and τ are small in comparison e.g. with the time scale t and the length scale x in an Eulerian coordinate system.

2 Atmospheric diffusion

There are many diffusion systems with a non-zero velocity field $\mathbf{v}$. So the atmospheric diffusion is influenced by different wind field aspects. The velocity vector field $\mathbf{v}$ (u, v, w) is splitted to its average $\mathbf{V}$ (ensemble mean) and an fluctuation part $\mathbf{v}'$

$$\mathbf{v} = \mathbf{V} + \mathbf{v}' \tag{12}$$

and the scalar concentration c in an adequate manner to

$$c = C + c' \tag{13}$$

The result of substituting (12) and (13) into (7) in combination with (10) and averaging by using the Reynolds postulates, e.g.

$$\overline{c'} = 0,\ \overline{C} = C,\ \text{etc.} \tag{14}$$

is

$$\frac{\partial c}{\partial t} + \text{div}\,(C\mathbf{V}) + \text{div}\,(\overline{c'\,\mathbf{v}'}) = \frac{S}{\beta} \tag{15}$$

In analogy to the kinetic theory of gases the average product is related to the average concentration gradient, compare with (9):

$$\overline{c'\mathbf{v}'} = -\mathbf{K}\ \text{grad}\ \mathbf{C} \tag{16}$$

where $\mathbf{K}$ is a 3x3 turbulent diffusivity tensor (Csanady, 1973). The tensor elements are determined from fluid flow analysis (Spaulding, 1976). In comparison with (9) the magnitude of $\mathbf{K}$ in horizontal direction is of order 10^4 cm^2/s. The atmospheric diffusion

$$\frac{\partial c}{\partial t} + \text{div}\,(C\mathbf{V}) - \text{div}\,(\mathbf{K}\ \text{grad}\ C) = \frac{S}{\beta} \tag{17}$$

involves no specific quality that refers to the emitted substances apart from source data and deposition velocities. It does not matter whether vanillin, ammonia or germs determine the atmospheric pollution provided that there is no relative motion caused by drag (Margolin, 1977). The calculation of immission is always the same. (17) can be solved generally by numerical methods only.

The numerical solution contains errors by numerical diffusion. Sklarew has shown that these effects can be avoided by introduction of an effective velocity (Hotchkiss, 1972)

$$\mathbf{V}'' = \mathbf{V} - \mathbf{K}\frac{\nabla C}{C} \tag{18}$$

The handling of problems works well when ∇C is resolved by a sufficient number of cells. In regions of strong flow distortion and poor resolution errors in the concentration distribution are obtained. The modeling of source constellation must be taken into account with care. In the following figures there is high resolution in the vicinity of buildings. Even the emitted mass flow is produced partly with vortex structures of the flow field in the stack outlet.

An alternative approach to (18) is to replace the diffusion velocity - **K** ∇C/C by a random velocity $\mathbf{V_R}$ (Zanetti, 1990) :

$$\mathbf{V_R} = -\mathbf{K}\frac{\nabla C}{C} \tag{19}$$

The sum of **V** and $\mathbf{V_R}$ is the total equivalent transport velocity.

$$\mathbf{V}'' = \mathbf{V} + \mathbf{V_R} \tag{20}$$

The diffusion equation reduces to the form

$$\frac{\partial c}{\partial t} - \text{div}\,(C\mathbf{V}'') = \frac{S}{\beta} \tag{21}$$

The original problem of turbulent atmospheric diffusion is transformed into one describing the advective change of fluid density C in a compressible fluid moving with the fictitious velocity field **V"** (Sklarew, 1971). The original boundary conditions have to be transformed. The physical space is divided into cells of a fixed Eulerian spatial grid and the particles carry pollution from cell to cell forced by the fictitious velocity field. This velocity field is not a solenoidal one, that means that the condition of conservation of mass is not fulfilled. So particles will move to maintain balance of mass. An uneven distribution of particles determines the average cell concentration.

There exists a hybrid method of Eulerian and Lagrangian techniques to solve the atmospheric diffusion equation. It is called **P**article-**I**n-**C**ell making use of the **K**-theory approximations ("**PICK**"). The Eulerian grid size determines the maximum spatial resolution. It is used for winds and values of concentration. Phenomena within grid size like point emissions can be represented by particles. The Lagrange approach for the modeling of dispersion is based on the calculation of the spatial trajectories of virtual particles moved with random velocity $\mathbf{V_R}$ (Janicke, 2001).

3 Significance of the flow field

Air pollution modelling is in general based on observations and theories of the surface layer (Nieuwstadt, 1981). The restriction to analytical solution of the governing equations in terms of known functions is lifted more and more by solving differential equations by digital computers directly. But the required resolutions with a great number of grid points damp the optimism. Turbulent flow contains eddy sizes from magnitude of 500 m to 1 mm. With respect to the conservation of momentum in a range of 10 km length, width and height 10^{18} grid points would be needed. But the resources are limited.

Figure 1 shows the dispersion of ammonia in the narrow surrounding of animal houses. Roughly 1.4 million cells are needed for this simulation. The Eulerian model technique is used. The pressure distribution around the buildings and the turbulence induced mechanically determine the distribution of ammonia, odour etc. It is a typical situation in a village (Krause, 2002).

In figure 2 the air motion around a broiler house is visualized by trajectories. It is intended to build a broiler house 60 m eastward from a little forest. A small valley lies between forest and broiler house. The trajectories show a complex velocity field. Here we need no surface layer contemplation, we need a detailed velocity field. Without a real velocity background Eulerian and Lagrangian modeling are senseless.

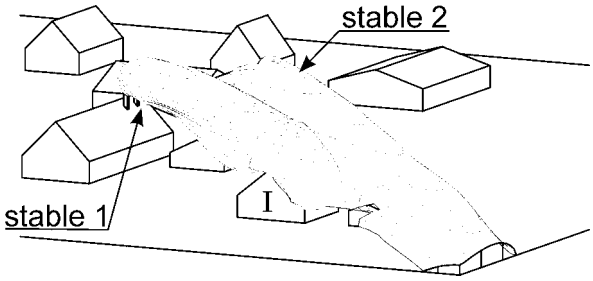

Figure 1:
Two stables emit air at low height over roof. The ammonia concentration amounts to 10 ppm. 5,040 m^3/h are thrown out at the front stable at every flue with a velocity of w_0 = 5.6 m/s, at back stable there are 4,224 m^3/h with a velocity of w_0 = 4.7 m/s. The shell areas of equal concentrations refer to C_0 = 0.14 ppm. The wind speed is U_{10} = 3 m/s at a height of 10 m. The puff is deflected. The building at the immission point I is coated by the ammonia puff.

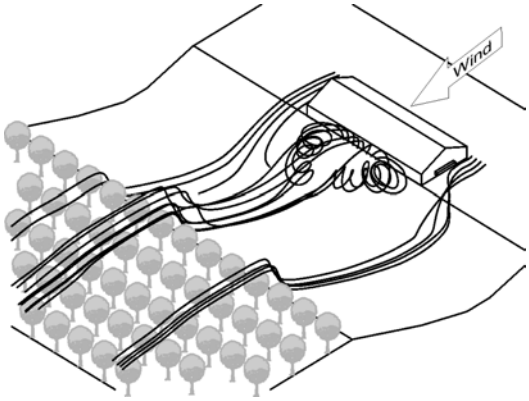

Figure 2:
Particle pathes (trajectories) around a broiler house. The wind blows from the right side perpendicular to the ridge line. On the lee side, that means in the shelter area behind the building, we observe strong turbulences that are mechanically induced. The stable air is thrown out at the gable in front of the perspective picture. This fact causes no symmetrical flow around the stable. A small deflection takes place to the gable in front. This is a typical example of obstacle influenced releases by near ground sources of agricultural animal production. It makes a great difference whether the stable air is emitted at the roof or at the gable. The difference is given by the initial distribution of ammonia concentration: by roof release we have so-called point sources with high density of ammonia, by near ground release we have so-called area sources with a great effect on the dilution when the air is ejected to the ground. The ground acts like a crash plate with respect to the mixing of stable air with the air in the surrounding.

The importance of a real velocity field is demonstrated by the turbulent viscosity at ground (figure 3) and at a small distance above ground (figure 4) induced by flow separation. The consequences are expressed by quite different ammonia distributions with respect to the forest (Krause, 2001). Figure 5 shows the broad ammonia puff around the broiler house with the isoarea of 10^3 µg/m^3. If the ammonia is exhausted vertically at the gable (figure 6) the forest is loaded hundred times higher then in the first case. Nevertheless, the administration decided to eject the stable air at the gable in vertical direction.

Figure 7 shows a so-called wind induced source, an outdoor climate stable (Krause, 2001). It is a very sensible system with respect to wind flow. We must learn to develop a feeling to the special problems of agricultural production. Crude simulation programs are misleading.

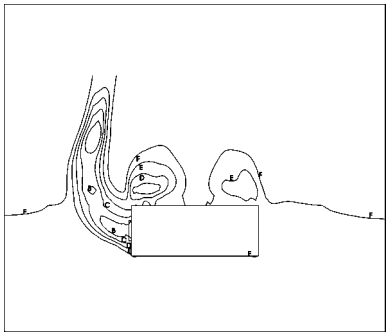

Figure 3:
The flow around the broiler house causes turbulence structures, top view of figure 2. The isolines of turbulent viscosity μ_T are pointed out at a height of 0.5 m above ground. The letters refer to the following values μ_T in 10-2 Pa s (= N s/m²)
A = 6.1, B = 5.08, C = 4.07, D = 3.05, E = 2.04, F = 1.02. In comparison the dynamic viscosity µ of air at 10 °C amounts to 1,81 10^{-5} Pa s. At the outlet on the left side we have a great production of vortices

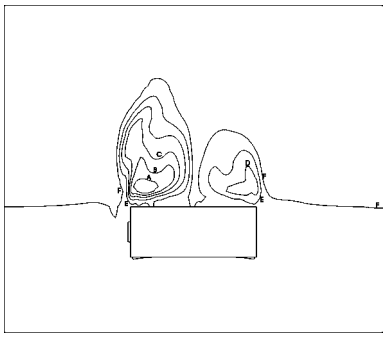

Figure 4:
The isolines with the same level of figure 3 are found in the shelter of the broiler house, here at a height of 3 m above ground

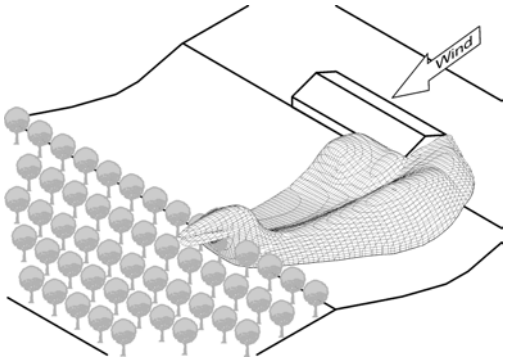

Figure 5:
Expansion of ammonia at exhaust on the gable of a broiler house. Perspective presentation of the isoarea of 10^3 µg/m^3. The exhaust air is blown out downwards on the gable wall in front. The concentration of the outer mantle of the cloud of ammonia shows a dilution of 0.001 compared to the outlet concentration

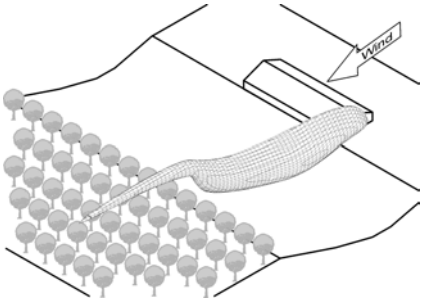

Figure 6:
Perspective view of the visible made cloud of ammonia induced by the emission from the broiler house and the windfield. The exhaust air is blown out upwards on the gable wall in front. The concentration of the outer mantle from the cloud of ammonia shows a dilution of 0.1 compared to the outlet concentration. The wind velocity has a magnitude of 2.5 m/s in a height of 10 m

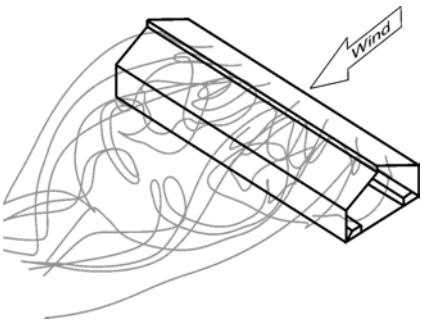

Figure 7:
Perspective presentation of an outdoor climate stable with shed roof. All length sides are covered with wind break nets. The opening in the ceiling can be closed or covered with a wind break net, too. The wind flows in optimal manner vertically to a side wall with a velocity of 4 m/s in a height of 10 m. The vertical flow is distributed as parabola profile. Different trajectories are shown beginning in the windward opening. The shed roof is open

4 Estimation of immission loads

To have confidence in simulation techniques does not prevent from plausible controls of results. In the VDI guidelines "Emission Control - Livestock Management - Pigs" (VDI 3471) and "Emission Control - Livestock Management - Hens" (VDI 3472) the so-called odour threshold distances were derived empirically. This is the distance at which - on approaching the facility - a facility-typical smell is first perceived or identified (Schirz, 1989). The distance r is described by

$$r = 48{,}69 \; M_{T,eq}{}^{1/3} \tag{22}$$

for optimal equipment. $M_{T,eq}$ is the odour-relevant total livestock mass value. At the distance r the source odour concentration C_0 is deluted to the threshold concentration C_S = 1 OU/m^3 and below:

$$q = \frac{C_0}{C_S} \tag{23}$$

The factor q alters from q = 350 in the surrounding of piggeries to q = 80 in the surrounding of cattle houses (Schirz, 1989). Improvements of the olfactometry measurement double the q-values nearly (Brose, 2001). The dilution factor q is valid not only for odour but also for ammonia, germs etc. The consequence is explained by the example of ammonia emission from swine houses. The average emission factor E (Umweltbundesamt, 2002) is

$$E = 3 \frac{\text{kg}}{\text{year animal} - \text{place}} \tag{24}$$

With an average pig mass of 70 kg and the Live mass Unit LU (= 500 kg) F is transformed to

$$E = 2.45 \frac{\text{g}}{\text{h LU}} \tag{25}$$

With a mean specific volume rate (Pedersen, 1998) of 300 m^3/(h LU) the source concentration is determined to

$$C_{0,NH_3} = 8.2 \frac{\text{mg}}{\text{m}^3} \tag{26}$$

Applying to (23) with q = 2 · 350 = 700 the ammonia immission concentration is

$$C_{NH_3} = \frac{C_{0,NH_3}}{q} = 11.7 \frac{\mu\text{g}}{\text{m}^3} \tag{27}$$

neglecting deposition. Tab. 1 shows the values for different livestock systems. The immission concentration must be compared with threshold values.

Doing so several aspects are worth to be mentioned. The natural background concentration of ammonia is about 4 µg/m^3, in rural areas 14 µg/m^3. Therefore it is a highly risky undertaking to demand threshold values below those values at the odour threshold distance, see tab. 1. On the other hand the question arises whether the factor q in (23) is of correct magnitude; perhaps it is to small and there is no ammonia problem at the odour threshold distance. With regard to germs we have the same situation as in the case of endotoxin. There are no known threshold values. All these statements base on the assumption of the atmospheric diffusion equation that the same dilution factor q is valid for all airborne gaseous substances from animal houses.

Table 1:
Immission concentration of different pollutant at the odour threshold distance. Though it is allowed to build a house or to live nearby animal houses, it may be forbidden with respect to ammonia to arrange a sensible ecosystem, because the threshold value of 10 µg/m^3 is exceeded. On the other hand the immission concentration stayed 93 per cent below the odour threshold value of 1 mg/m^3. In a piggery the mass of 60 LU causes an ammonia emission of 1.29 Mg/Jahr. After TA Luft the minimum distance X_{min} is calculated to $X_{min} = (41{,}668 \cdot 1.29)^{0,5} = 232$ m. The odour threshold distance according to (22) is 166 m

Species	factor q	mean specific volume rate in m^3/(h LU)	ammonia			endotoxin		
			emission factor E in g/(h LU)	Source concentration C_0 in mg/ m^3	**immission concentration C in µg/ m^3**	mean respirable emission in µg/(h LU)	Source concentration C_0 in ng/ m^3	**immission concentration C in ng/ m^3**
cattle	160	150	1.7	11.3	70.6	1.2	8	0.05
pig	700	300	2.45	8.2	11.7	5.9	20	0.03
poultry	760	750	2.8	3.7	4.9	42.7	4	0.005

5 Conclusion

Better solutions are enemies of good ones. Indeed the Gaussian plume model is a first approach to estimate immission loads by analytical solution of the atmospheric diffusion equation. The development of efficient grid generators, the improvement of computational methods and the progress in computer based solutions broaden the possibilities to elaborate more precise statements to environmental problems. The update of the TA Luft reveals a great dilemma: to improve the immission prognosis by substituting the Gaussian model by a Lagrangian one is no real progress because of the lack of realistic flow field calculation. From this point of view the Lagrangian model AUSTAL2000 with a meteorological flow background of the planetary boundary layer does not realize the situation of agricultural production with complex flow situation in the surrounding of animal stables. Furthermore there is a great lack of ammonia immission data of the surrounding of animal houses to validate dispersion models.

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Modelling PM 2.5 concentrations over Europe: the role of agriculture

M. Schaap [1], M. Van Loon [2], A. Visschedijk [2], F.J. Dentener [3] and P.J.H. Builtjes [1,2]

Abstract

Emissions from agricultural activities influence the concentration of fine particulate matter on a European scale. Ammonia plays a key role in the formation of secondary inorganic PM by neutralising acid gases. In this paper the LOTOS model is described, which has been used to simulate the fine inorganic particulate mass over Europe. In addition, concentrations fields due to primary emitted particles were calculated.

Sulphate was neutralised by ammonia over the whole domain. Excess ammonia is then available for the formation of ammonium nitrate, which was found to be present in similar amounts as sulphate during winter. In summer nitrate seems to be limited to north western Europe and the Po valley in Italy.

The model is able to reproduce spatial and seasonal features as have been observed in reality. Agreement between model and measurements are favourable at continental sites in north western and central Europe. However, in coastal regions nitrate concentrations are underestimated.

During winter the formation of ammonium nitrate appears to be sensitive to the availability of nitrate. The sensitivity to ammonia seems to be less, when only the partitioning of nitrate between the particulate and the gaseous phase is studied. However, changing the ammonia emission by 25 % shows a non-linear response with 10 – 50 % change in the nitrate concentration over large areas in Europe. Primary emissions of fine particles due to agricultural activities do not contribute a large fraction to PM levels in Europe.

Keywords: Particulate matter, Nitrate, Sulphate, Ammonium, Primary emissions, Modelling

1 Introduction

Particulate matter plays an important role in environ-mental issues as health, acidification, eutrophication and climate change. PM has been associated with adverse health effects such as respiratory deceases and mortality (Pope et al, 1995). A large fraction of fine PM consists of sulphate, nitrate and ammonium, which cause acidification and eutrophication when deposited to the ground. Due to their lifetime, in the order of a week, fine particles can be transported over large distances and cause effects far away from the source regions of their precursors. Sub-micron particles (d<1 µm) effectively scatter light back into space and therefore influence the radiation budget of the earth directly. Indirectly, fine particles influence the properties and, therewith, the effective albedo of clouds.

Ammonia is the most abundant base gas in the atmosphere and plays an important role in the formation of secondary inorganic particulate matter. Ammonia acts as a neutralising agent for particulate sulphuric acid:

$$NH_3 + H_2SO_4 \rightarrow NH_4HSO_4 \quad (1)$$

$$NH_3 + NH_4HSO_4 \rightarrow (NH_4)_2SO_4 \quad (2)$$

When sulphuric acid is completely neutralised excess ammonia (further denoted as free ammonia) can react with gaseous nitric acid under formation of the particulate semi volatile ammonium nitrate:

$$NH_3 + HNO_3 \rightarrow NH_4NO_3 \quad (3)$$

The gas aerosol partitioning of nitric acid has been shown to be strongly dependent on the availability of its precursor gasses and ambient conditions (Ansari and Pandis, 1998). Sensitivity studies with equilibrium models have shown that under conditions where ammonia limits the formation of nitrate, the total secondary inorganic mass behaves nonlinearly with changing ammonia availability. A decline of sulphate concentrations under those conditions can be largely compensated by a subsequent rise in nitrate levels (West et al, 1999). In an ammonia rich environment the total mass of nitrate, sulphate and ammonium declines linearly with nitrate availability and sulphate concentration. In order to assess the

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effects of present and future abatement strategies, it is important to understand the formation and loss processes of inorganic PM and the sensitivities of the system.

Schaap et al [2002] critically assessed the present measured contribution of nitrate to the aerosol burden in Europe, north of the Alps. The reason for the exclusion of southern Europe was that only two data points were available, against 31 north of the Alps. In winter large contributions of nitrate were found (see figure 1), especially in western Europe where nitrate concentrations exceed those of sulphate. Nitric acid concentrations were found to be about one tenth of those of particulate nitrate. Except for the Netherlands a distinct seasonal variation was found in the nitrate data with maximum values in winter and low values in summer. Therefore, it seems realistic that different regimes for the formation of nitrate are present in Europe, as function of region (ammonia emission density) and season. As a consequence, the effectiveness of abatement strategies for NO_x, SO_x and NH_3 could be different in those regions.

Ammonia emissions are mainly caused by agricultural activities. Additionally, agricultural activities cause direct emission of particles in the atmosphere. Hence, emissions from agriculture influence the levels of particulate matter on a European scale. We evaluate the contribution of ammonium nitrate and primary emitted particles to total particulate mass over Europe, using the 3D atmospheric-chemistry transport model LOTOS.

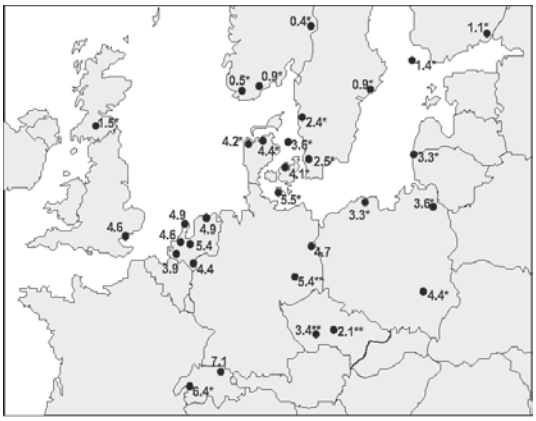

Figure 1:
Average particulate nitrate concentration during winter (Okt- Mar) as compiled by Schaap et al. (2002) (* total nitrate data corrected for nitric acid).

2 The LOTOS model

2.1 Model description

The LOTOS model is a three-dimensional chemistry transport model of intermediate complexity covering Europe. The idea behind the model is that it should contain all relevant processes (explicitly or in a parameterised form) in such a way that hour by hour calculations over periods of years are feasible. LOTOS was originally developed for ozone modelling, using CBM-IV as chemical scheme. In the last few years the model has been extended for aerosols. A general and detailed description of the model can be found in (Schaap et al., 2002). Below, some relevant details on the model concept and the modelling of aerosols within LOTOS are given.

The horizontal resolution of the model is 0.5 by 0.25 degrees lon-lat. In the vertical the boundary layer and the lower free troposphere are represented by concept of dynamical layers: the depths of the three layers depend on the (in time and space varying) mixing layer height. The first layer represents the mixing layer, the other two (reservoir) layers are equally distributed over the rest of the vertical domain up to 3.5 Km. The model contains a parameterised surface layer. The diagnostic meteorological input for LOTOS (winds, temperature, moisture, mixing layer height, etc) is prepared by the Free University of Berlin. Gridded emission data of SO_x, NO_x, VOC and NH_3 are calculated by TNO for the base year 1995.

Emission or deposition of ammonia depends on the so-called compensation point, which refers to the situation that the ammonia concentration in air is in equilibrium with that in the vegetation and soil. Assessing the compensation point of ammonia is not yet possible for many surfaces (Asman,2001). Therefore emission and deposition are described separately. The annual average ammonia emissions used in this study are shown in figure 2. The seasonal variation in ammonia emissions is modelled based on data representative for the Netherlands (Duyzer, pers. communication). Due to these emissions there is a large vertical gradient of ammonia concentrations in the source areas with highest concentrations near the surface. However, in our model the emissions are vertically mixed over the mixing layer. We will therefore underestimate the dry deposition of ammonia in these areas. Following Dentener et al (1994) we lowered the emissions by 25 % to account for this effect (so called “direct dry deposition”).

Sulphate is formed in the gaseous phase as well as in the liquid phase. The oxidation of sulphur dioxide by the OH radical is represented in the gas phase reaction mechanism CBM-IV. Another important

oxidation pathway is the formation of sulphate in clouds. Since our model considers a simplified hydrological cycle, we represented it with a first order reaction constant that varies with cloud cover and relative humidity, similar to the approach followed in EUROS (Matthijsen et al. 2002). Nitric acid is also formed via two distinct pathways, homogeneous in the gas phase (CBM-IV) and heterogeneous on the surface of aerosols. Both processes are treated explicitly in the model.

The equilibrium between ammonia, nitric acid and ammonium nitrate is very sensitive to ambient conditions and is calculated using a modified version of the MARS system (Ackermann et al., 1995). At this moment the LOTOS model does not incorporate sea salt. Therefore, caution is advised interpreting the results of the model for marine conditions (Zhang et al, 2000).

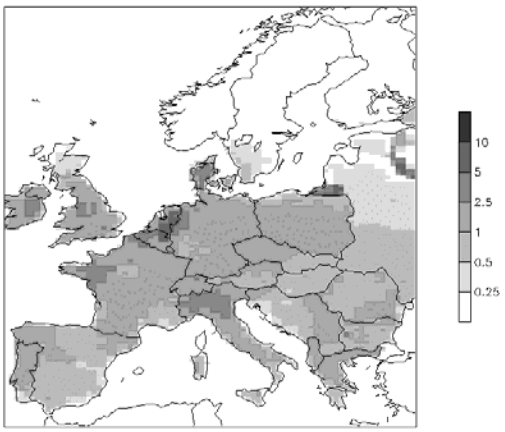

Figure 2:
Gridded annual average ammonia emissions (*1000 Ktonnes/yr) for 1995

2.2 Primary emissions

A primary emission database of fine particulate matter has been composed within the CEPMEIP project at TNO (TNO, 2001). CEPMEIP contains sectoral information on for instance industry, traffic and agriculture. In short, the primary particulate matter emissions from agriculture in CEPMEIP have been estimated on a country basis, based on international statistics for animal populations, emission factors for different animals and a distribution map. CEPMEIP only includes PM emissions from animal farming and agricultural waste burning.

CEPMEIP regards four animal types: chicken, other poultry, cattle and pigs. Animal populations have been taken from the FAO database. Emission factors are based on an international state of the art review by Takai et al. (1998) and are the same for all countries. The fraction PM10 and PM2.5 is estimated based on various measurement results in the Netherlands and comparisons with data for total, inhalable and respirable dust. Emissions from animals have been spatially distributed according to van de Velde et al (1994)

Emissions from agricultural waste burning have been based on activity values and distribution patterns from the EDGAR V3.2 database (Olivier et al, 2001). The emission factor for PM2.5 from agricultural waste burning has been based on a TNO review of the work by M.O. Andreae and Jan Hulkotte, undertaken within the framework of CEPMEIP. The gridded primary emissions of PM2.5 are shown for north western Europe in figure 3.

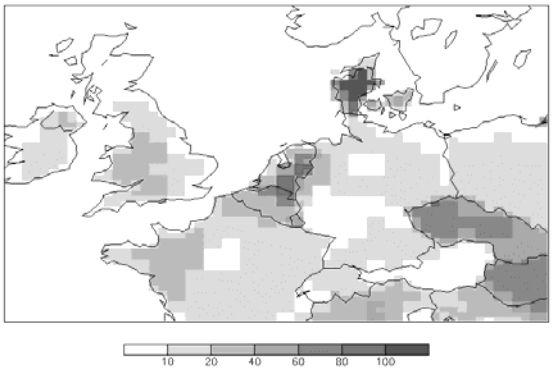

Figure 3:
Primary emissions in north western Europe of PM2.5 by agriculture (tonnes/yr)

3 Results

The LOTOS model has been applied from January to November, 1995.

3.1 Secondary ions

In figure 4 the nitrate field for June to August is shown. In these summer months nitrate is confined to western Europe with maximum concentrations over the Netherlands. Other regions with high concentrations can be identified over northern Italy, England and the Balkans. These areas are characterised by high ammonia emissions. The ambient conditions (high temperatures) in eastern and southern Europe do not favor ammonium nitrate formation. Therefore, sulphate is much more abundant in the summer. A band of high sulphate concentrations is calculated over western Europe to the Balkans with maximum concentrations in Poland and south eastern Europe. The sulphate was almost

completely neutralised by ammonium in the model domain.

In the winter nitrate shows a different behaviour than in the summer (see figure 4b). The calculated nitrate concentration field shows a large area of high nitrate concentrations over western and central Europe. The high nitrate concentrations can be explained by the much higher stability of ammonium nitrate at low ambient temperatures. In western Europe the calculated nitrate concentrations exceed those of sulphate. In eastern Europe nitrate concentrations are slightly lower than those of sulphate.

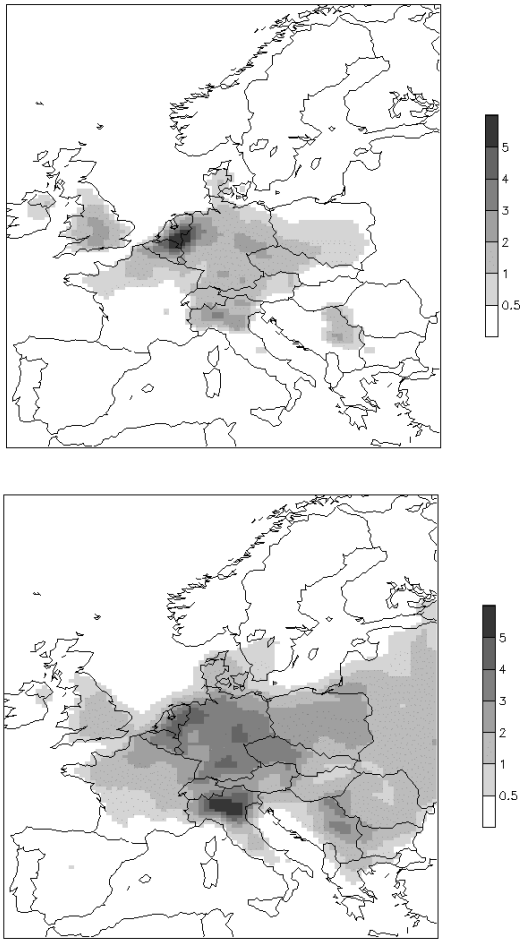

Figure 4:
Aerosol nitrate field (µg/m^3) for a) summer (Jun-Aug) and b) winter (Oct-Mar)

The results of LOTOS have been extensively compared with measured data for the whole of 1995 (Schaap et al, 2002). Here we summarise the process and its results for the selected months. Verification of the model results is only than possible when reliable data are available. For sulphate and sulphur dioxide a large database of reliable data exists, e.g. EMEP (EMEP, 2002). Measurements of nitrate are sparse and prone to artefacts because of the volatility of ammonium nitrate and the reactivity of nitric acid. We use the database selected and evaluated by Schaap et al (2002) to verify our model results. Reliable data are only obtained with devices that remove nitric acid prior to aerosol sampling and stabilise ammonium nitrate against evaporation, e.g. denuder filter combinations. During summer evaporation of ammonium nitrate causes negative artefacts on inert filters. Positive artefacts are possible by adsorption of nitric acid to previous collected aerosol. In Europe north of the Alps evaporation is thought to dominate over the adsorption of nitric acid. Hence, the data obtained are minimum values. In winter both evaporation and adsorption of nitric acid is thought to be small. Data from cellulose filters are interpreted as to be total nitrate, the sum of aerosol nitrate and gaseous nitric acid.

In figure 5 the calculated annual averages are compared to measured data in order to assess the basic performance of the model. Given the mentioned possible artefacts we chose to compare the modelled aerosol nitrate concentrations to data acquired with denuder techniques for the whole year and to those acquired with inert filters for the winter only.

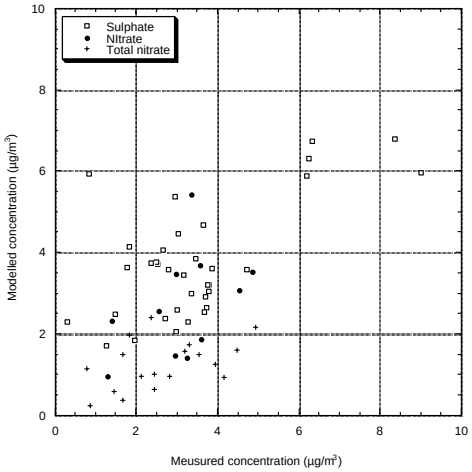

Figure 5:
Comparison between measured and modelled concentrations of sulphate and (total) nitrate.

The sulphate concentration field over Europe is captured quite well, as can be seen from the favourable comparison with measurements. This is necessary in order to model the available ammonia for ammonium nitrate formation correctly. Overall, the modelled nitrate concentrations are lower than the measured concentrations. This feature is clearly seen for the total nitrate sites, which are mostly located in coastal regions. The agreement between modelled and measured values is much better for the continental

sites, which are mainly located in central and north western Europe.

As indicated above, the performance of the model is best for the continental sites in western Europe, e.g. Melpitz, Muencheberg and Vredepeel. In figure 6 and 7 the daily variation of nitrate and sulphate is shown for Muencheberg, which is located near Berlin, and Vredepeel, the Netherlands.

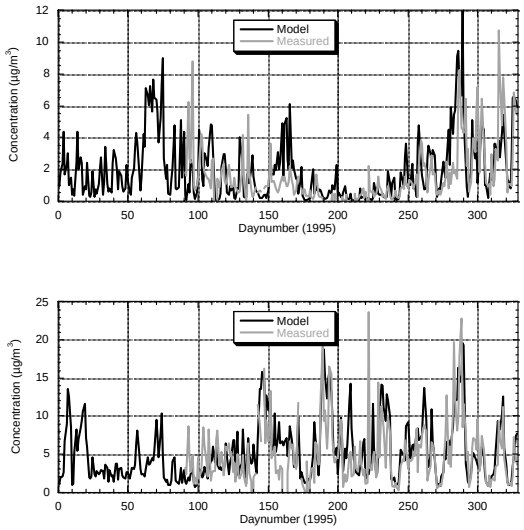

Figure 6:
Daily averaged measured and modelled concentrations of nitrate and sulphate (upper and lower panel, respectively) (µg/m^3) for Muencheberg, Germany

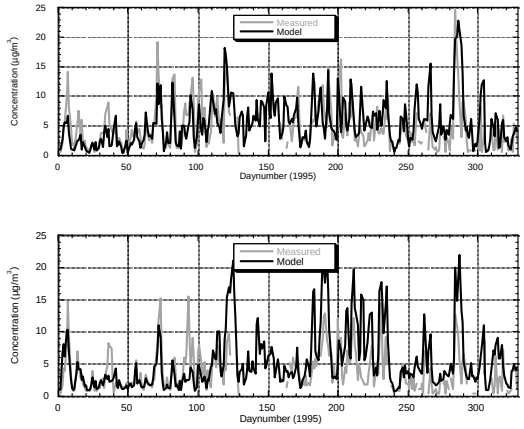

Figure 7:
Daily averaged measured and modelled concentrations of nitrate and sulphate (upper and lower panel, respectively) (µg/m^3) for Vredepeel, the Netherlands

3.2 Primary PM2.5

The annual average concentration field for primary emitted fine particulate matter (PM2.5) due to agricultural activities is shown in figure 8. Concentrations range from 0.22 µg/m^3 in Hungary to nearly zero in remote locations as northern Scandinavia. Since the primary particles are treated as chemically inert species the concentration field shows a large similarity with the emission field. At present it is not possible to verify these results against measurements. However, relative to the concentration of ammonium nitrate the contribution of primary particles is low.

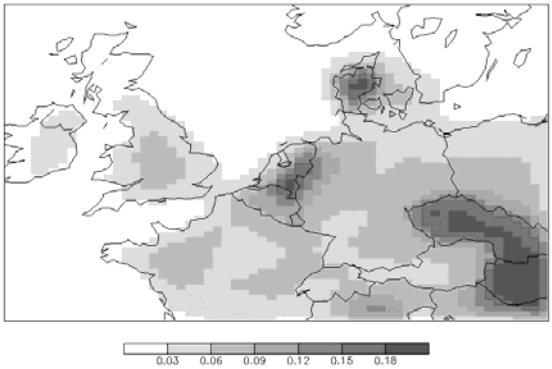

Figure 8:
Modelled contribution (µg/m^3) of primary emissions of agriculture to the PM2.5 burden during 1995 over north western Europe.

3.3 Wintersmog

Highest concentrations of particulate matter as a total and (ammonium) nitrate occur during the winter half of the year. We therefore focus on winter to address the formation mechanism of the nitrate in the model.

In order to assess the regime, in which the equilibrium between the gas and particulate phase is, the change in the nitrate concentration was determined for small changes in the concentration of the precursor gases. The hourly output values of total ammonia and total nitrate were raised and lowered by 5 % in order to define the sensitivity to these parameters. In figure 9 the sensitivity for changes in the total nitrate and total ammonia concentration are shown for January. For a large part of the domain, i.e. the continental regions, the sensitivity is large for changes in nitrate concentrations, more than 75 % of the added or subtracted nitrate is found in or lost from the aerosol phase. The sensitivity for ammonia seems less than for nitrate, and is complementary to that of nitrate.

This exercise only takes into account the chemical equilibrium between the species involved. However, in reality the picture is much more complex. Changes in the available amount of ammonia for the formation

of nitrate, e.g. due to higher emissions, may be masked by other processes or feedback mechanisms. Therefore we have done a sensitivity run for January in order to see how the ammonium nitrate concentration field changes with a 25 % higher ammonia emission. The resulting change in nitrate concentrations is shown in figure 10. Raising the ammonia emissions by only 25 %, which is much less than the uncertainty associated with the emission data, causes nitrate concentrations to rise significantly, e.g. more than 10-20 % over most of Europe. In areas where ammonia emissions are highest, e.g. the Netherlands, the sensitivity to ammonia emissions is lowest, which can also be clearly seen in figure 9. In (south) eastern Europe nitrate levels are shown to be sensitive to ammonia emission changes. The large sensitivity to ammonia over the open sea is largely artificial: concentrations are low and nitric acid will react with sea salt rather than with ammonia.

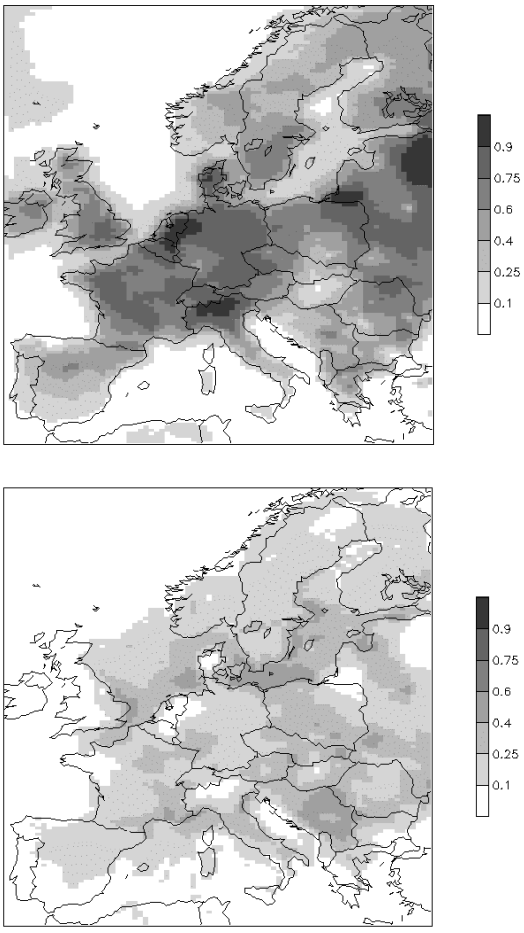

Figure 9:
The percentile change in nitrate concentration due to small changes in available nitrate (upper panel) and ammonia (lower panel) for January, 1995.

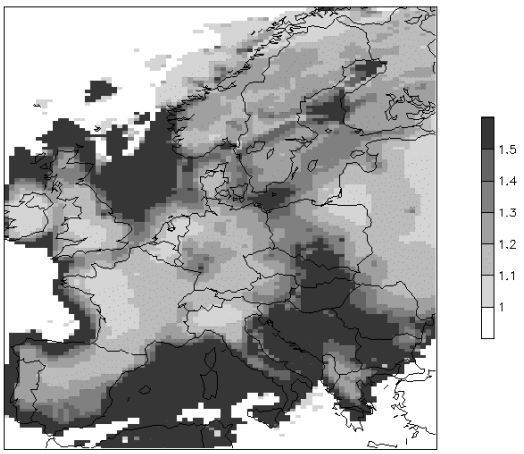

Figure 10:
Ratio of the average nitrate concentration using 25 % higher ammonia emission strength to that for the base case in January, 1995.

4 Discussion and conclusions

Emissions from agricultural activities influence the concentration of fine particulate matter on a European scale. Ammonia plays a key role in the formation of secondary inorganic PM by neutralising acid gases. In this paper the results of LOTOS are shown for 1995. It was found that the model underestimates nitrate levels over Europe, both in summer and in winter. The underestimation is mainly associated with stations in coastal regions. Several reasons could exist for the discrepancies found between the model and the measured data:

1. Emissions of ammonia are uncertain and due to the sensitivity of the model to ammonia concentrations these uncertainties have a large impact on the results. In addition, the treatment of the exchange of ammonia between the soil/biosphere and the atmosphere is still crudely represented in the model(s).
2. The model does not incorporate sea salt. Both fine and coarse mode sea salt stabilise nitrate. Neglecting sea salt in coastal and marine areas will cause underprediction of nitrate levels.
3. Uncertainties in the description of the equilibrium. The thermodynamic quantities used in the calculation of the equilibrium between nitric acid and ammonia are uncertain. These uncertainties influence the partitioning mostly at high ambient temperatures.

In north western Europe nitrate concentrations were found to be comparable to those of sulphate, throughout the whole year. In (south) eastern Europe a clear seasonal cycle can be observed. These findings are in agreement with measured data (Schaap et al, 2002). The model underestimates nitrate

concentrations in coastal regions. During winter the model is able to reproduce the spatial distribution as presented in figure 1. It was found that the formation of particulate nitrate was most sensitive to the availability of nitric acid. The partitioning of nitrate between the gaseous and particulate phase appears to be less sensitive for the availability of ammonia. However, a sensitivity run with 25 % higher ammonia emissions showed that nitrate levels increased by at least 10 – 20 %. Over large areas the concentrations raised by more than 25 %, a clear indication that the formation of ammonium nitrate is strongly non-linear. Due to the volatility of ammonium nitrate these sensitivities are a function of several parameters and should be addressed further in the future.

Primary emitted particles due to agricultural emissions contribute an order of magnitude less mass to total PM than secondary formed particles. However, it should be kept in mind that these emissions only take into account the emissions by animal farming and agricultural waste burning Exhaust emissions from stationary sources and motorised vehicles used in agriculture are not included and may be significantly higher. We could not separate the contribution of these emissions because they are included under different sectors, in cause of motorised vehicles the (off road) traffic emissions.

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Chemical coupling between ammonia, acid gases, and fine particles

Bok Haeng Baek and Viney P. Aneja[1]

Abstract

Transfer between gas and particle phases is time dependent and not instantaneous, and should be simulated with time-dependent growth equations. Under certain atmospheric conditions, the volatile inorganic compound aerosols may not be in equilibrium with their gas phases due to the lack of transport time needed to be in chemical equilibrium.

In this present study, the equilibrium time constant will be determined based on kinetic rate constants and the observed inorganic components of atmospheric aerosols. The predicted equilibrium state chemical compositions in the particle by thermodynamic equilibrium model (EQUISOLV II) will be compared with the observed chemical compositions at the experimental site.

Introduction

A particle in the atmosphere may grow in size by mass transfer of gas to its surface followed by conversion of the gas at the surface by condensation, deposition, chemical reaction, dissolution, or dissociation processes.[1] The chemical composition of an atmospheric particle may be inorganic and/or organic aerosol such as ammonium, sodium, sulfate, nitrate, chloride, water, soil dust, elementary carbon, and organic carbon. Inorganic aerosol salts, comprise 25-50% of dry total fine aerosol mass.[2,3] The particles consist of an aqueous phase at high relative humidity, one or more solid phases at low relative humidity, and both aqueous and solid phases at intermediate relative humidity. Especially, chemical reactions between ammonia with nitric acid and hydrochloric acid to form ammonium nitrate and ammonium chloride are reversible processes in the atmosphere due to higher volatility. The assumption of thermodynamic equilibrium has been employed to partition the volatile compounds between the gas and particle phases.[4,5,6,7,8,9,10] The interaction between the gas and particle surface are not easily resolved and vary amongest particles because many of the processes are tightly coupled. If the ambient gas phase concentrations exceed those over particle surface, the gases diffuse into and condense on the particles.

Over the past decades, considerable effort has been directed towards the understanding of the physical and chemical properties of inorganic aerosols, while several inorganic aerosol thermodynamic models have been developed. All of these models assumed that internally mixed particles, i.e., all particles simulated in a given particle size range have the same chemical composition, and that thermodynamic equilibrium exists between the gas and particulate phases for the volatile compounds. Thus, particles of different size, consisting of primarily ammonium, sulfate, nitrate, and chloride should have the same composition ratios, if they are in equilibrium between the gas and particle phases. This assumption has been applied to the thermodynamic equilibrium models. Zhang et al., (2000) also reviewed the similarities and differences in simulation results predicted by five thermodynamic equilibrium models, such as MARS-A[9], SEQUILIB[12], SCAPE2[13,14,15,16], AIM2[17,18,19,20], and EQUISOLV

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II[21].[11] They recommended that the EQUISOLV II could be applied to simulate particulate matter for any conditions. Moreover, they suggested application of the EQUISOLV II for three-dimensional particulate matter modeling studies.

To attain high computational efficiency, assumptions have been made to simplify the problem and allow use of the equilibrium models in Eulerian atmospheric chemical transport models. Three-D models capable of tracking aerosol dynamics, such as transport of gases to particles, particle formation, and size distribution, have been combined to simulate the behavior of organic and inorganic aerosols. Although limited comparisons among equilibrium models have been conducted[22,14], no detailed comparisons between observations and predicted aerosol behavior have been performed due to limited availability of such measurements[21,23].

The kinetic chemical rate constants of ammonia with gaseous sulfuric acid, gaseous nitric acid, and gaseous hydrochloric acid in the atmosphere have been estimated.[24] The objective of the present study is to estimate the equilibrium time constant for transfer between gas to particle phase of ammonium nitrate and ammonium chloride, and to compare the observed data with those predicted by a thermodynamic equilibrium model, EQUISOLV II which is recommended for any condition by Zhang et al., (2000).[11]

Measurements

The study area consisted of two measurement sites at a commercial hog farm in eastern North Carolina, dubbed North Farm (NF) site and South Farm (SF) site. The chosen study site is a 1200 sow farrow to finish operation in eastern North Carolina. The sampling site has a waste lagoon with a surface area of approximately 2.7 hectares (2.7×10^4 m^2), and animal housing units consisting of 9 finishing barns, 2 nursery barns, and 2 farrow barns. Figure 1 shows the prominent features of the operation, including monitoring sites. The NF site was located approximately 50 meters northeast of the swine waste storage and treatment lagoon, and SF site was located approximately 400 meters south-southwest of the waste lagoon. Samples were collected using the annular denuder systems (ADS) from April to July 1998 at NF site, and from April 1998 to March 1999 at SF site. The ADS system consisted of a cyclone separator to remove coarse particles (>2.5μm), which are likely to be alkaline, to prevent neutralization of acid gases and acid fine particles, two diffusion denuders in series coated with sodium carbonate and citric acid, respectively, and a filter pack containing a Teflon and a nylon filter (figure 2). The sampler inlets were approximately four meters above the ground. Sampling flow rates and intervals were controlled using mass flow controllers and electronic timers, respectively. The samplers were operated at a constant flow rate of 10.0 $l\ min^{-1}$. ADS measurements were derived from 12-hour integrated samples, representing daytime and nighttime averages.

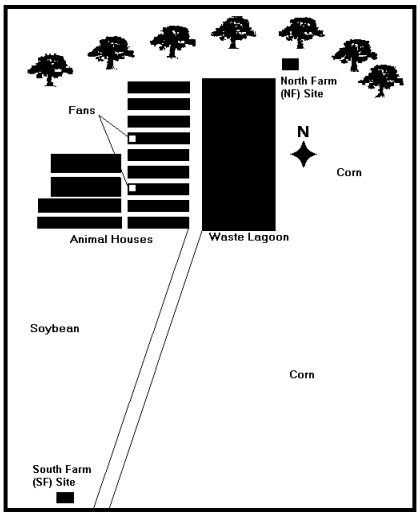

Figure 1:
Layout of sampling sites

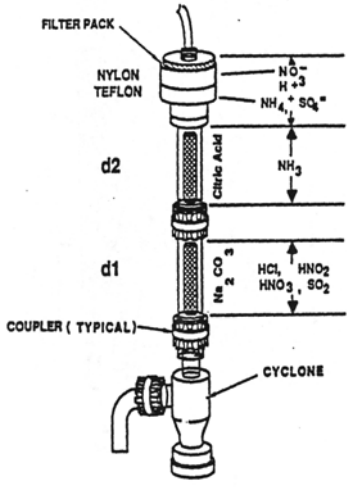

Figure 2:
Schematics of Annular Denuder System

The sodium carbonate denuders coated with 1% glycerine and 1% sodium carbonate ($NaCO_3$) in a 50% mixture of methanol and distilled water collects average gas concentrations of hydrogen chloride (HCl), nitrous acid (HONO), nitric acid (HNO_3), and sulfur dioxide (SO_2). The citric acid denuders were coated with 2% citric acid in a 50% mixture of

methanol and distilled water to capture ammonia (NH_3), which must be removed prior to reaching the filter pack. The filter packs contained both 47-mm membrane Teflon and nylon filters. The 2.0 μm pore size Teflon filter was used for collecting fine particle concentrations of ammonium (NH_4^+), chloride (Cl^-), nitrate (NO_3^-), and sulfate (SO_4^{2-}) ions in the atmosphere. The 1.0 μm pore size nylon filter next to the Teflon filter was installed to capture nitrate from the ammonium nitrate, which may have dissociated from the Teflon filter.

The denuder samples were extracted with deionized water and the Teflon filters were extracted in a heated sonic bath with 10^{-4} N perchloric acid in a 2% mixture of methanol and deionized water. The nylon filters were extracted in a heated sonic bath. The extracted samples were analyzed for chloride, nitrite, nitrate and sulfate by ion chromatography and for ammonium by colormetric flow injection analysis.

Meteorological parameters including wind direction (WD), wind speed (WS), and air temperature were measured at a tower near SF site at 2-m and 10-m elevations. Relative humidity was measured at 2-m. Solar radiation and precipitation were measured at ground level.

Input Data

Although wind directions fluctuated to some extent within each sampling period, it was possible to select data with a great predominance in the appropriate wind sector. Figure 3 summarizes the frequency of ammonia concentration regime by wind direction. During the measurement period, not only was the southwest wind direction dominant but also higher concentrations of ammonia occurred at the NF site, which was effected from the nearby waste lagoon. On the other hand, during the northeast wind direction, higher ammonia concentrations occurred at the SF site.

The measurement time period of ammonia, acid gases, and fine particle obtained by annular denuder system (ADS) were derived from 12-hour integrated samples, representing daytime and nighttime averages. Since accurate measurements of the acid gases, and fine particles in the atmosphere are difficult due to the technical limits of ADS even when sampling times in excess of a few hours are employed, it would appear that comparison between the NF and SF sites may not be discernible. Likewise, there are some other uncertainties about the measurements.

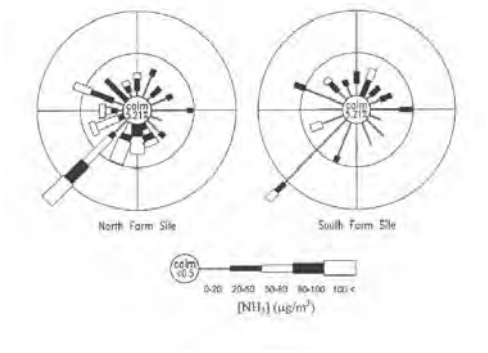

Figure 3:
The frequencies of ammonia concentration regime by wind direction.

Gaussian Dispersion Model

To consider additional ammonia contribution from lagoons and barns for this study, the ammonia flux was estimated by a Coupled Mass Transfer and Chemical Equilibrium Reaction Model. The normalized absolute difference between the observed and predicted ammonia flux from a swine lagoon is 39.72% on average.[25] The model predictions of flux indicate an exponential increase in ammonia flux with increasing lagoon temperature and pH, a constant increase with increasing lagoon Total Ammoniacal Nitrogen (TAN), and an increase with increasing wind speed. Input parameters for the ammonia flux model, such as lagoon pH 7.7(±0.06), TAN 603.3(±48.2) mg N/l, and lagoon temperature 24.7(±3.2) ^oC, were obtained from Aneja et al. (2000).[26] Ambient temperature, wind speed at 10 meters, and ambient ammonia concentration at this study site were required for ammonia flux emission. The average estimated ammonia emission from the waste lagoon was 5.38×10^2 μg NH_3/m^2/min during the measurement periods. The ammonia emission from barn is 3.09 kg NH_3/animal/yr.[27]

For conversion of the estimated ammonia emissions from a swine lagoon and barns to the contributed ammonia concentrations to the receptor, a Gaussian dispersion model is used. The model has been validated against experimental data due to experimental and theoretical input parameters, such as stability classes and effective wind speed.[28] A stability class was determined by the extended conditions of Pasquill Stability types based on hourly meteorological parameters.[29] Briggs' interpolation formulas[30] were used for assigning dispersion parameters, such as σ_y and σ_z. based on Pasquill stability classes.[29,31]

In this study, point sources were assigned to the exhaust fans of the animal housing units, and the waste lagoon was divided into 40 cells, acting as

individual point sources, rather than a single area source. This allowed a consistent modeling approach for both source types, and also allowed concentrations based on the influence of various fractions of the lagoon, depending upon the wind direction and short transport distances involved. A Gaussian dispersion model for estimating concentrations downwind of an area source which is divided into discrete multi-point sources located at the centers of grid cells has been used earlier.[32]

For the criteria data selection, wind direction dependent geometric parameters were computed for each point source. The fixed parameters are the north to south (a_i) and east to west (b_i) distances from each point source to the receptor, the resultant distance (r_i), the mean direction from which the wind is coming (Θ), and associated angle (α_i) , where *i* is the number of cells which contribute to receptor ammonia concentration. For separating the point sources, which contributed to the ammonia concentration at the receptor site, we compared the crosswind distance (y_i) to the estimated plume half-width ($2.15\sigma_y$) at the downwind distance from the center of the lagoon to the receptor site (SF). If the crosswind distance was less than the plume half-width ($2.15\sigma_y$), the point source was considered to contribute to the measured ammonia concentration at the receptor. The lagoon and barns were treated as an array of individual point sources, rather than a single area source. This allowed for the consideration of various fractions of the lagoon, depending on wind direction.[33]

The method of images is used to consider the reflecting surface assumption. This implies an image source below the surface to account for the increased concentration in the plume, and a perfectly reflecting surface, such that all mass emitted is contained within the plume, and therefore there is no deposition of material. However, considering the effects of dry deposition of ammonia gas on contribution of ammonia source to receptor, the partial reflecting assumption developed by Overcamp (1976)[34] is used. The contribution of ammonia source to a receptor is reduced by a factor (α), called the reflection coefficient. When α=1, the lower boundary can be described by the perfectly reflecting assumption, and the upper boundary by the non-reflecting assumption (α=0). In this partial reflection model, the mean concentration is give by Equation 1.1 for steady-state condition, and the express for the reflection coefficient at z = 0 is given by Equation 1.2, where *i* is the number of cells which contribute to receptor ammonia concentration.

$$\overline{C}_i(x,y,z,t) = \frac{Q_{NH_3}}{2\pi \bar{u}\sigma_{yi}\sigma_{zi}} \exp\left(-\frac{y_i^2}{2\sigma^2{}_{yi}}\right)\left\{\exp\left[-\frac{(z-H)^2}{2\sigma^2{}_{zi}}\right] + \alpha(x,z)\exp\left[-\frac{(z+H)^2}{2\sigma^2{}_{zi}}\right]\right\} \tag{1.1}$$

$$\alpha_o(x) = 1 - \frac{2v_d}{\left(v_d + \frac{\bar{u}H}{\sigma_z}\frac{d\sigma_z}{dx}\right)} \tag{1.2}$$

The average concentration at the downwind receptor of the source is indicated by C_i ; u is the effective wind speed called the transport speed of the plume ; Q_{NH3} is the emission source strength of ammonia ; σ_y and σ_z are the lateral and vertical dispersion parameters, respectively ; α is the reflection coefficient ; x_i is the along-wind distance from the source ; y_i is the crosswind distance from the center line of the plume to the receptor ; z is the height of the receptor ; H is the height of the emission source, and v_d is the dry deposition of NH_3. In this study, an NH_3 dry deposition value of 16 mm/sec on the natural meadow cut based on Sutton et al., (1993)[35] is used for this study.

For estimating contributions of ammonia emission from the swine lagoon nd barns to the receptor, a Guassian dispersion model has been executed based on every hourly NH_3 emission rate and meteorological input data for 12 hours due to the availability of 12 hours integrated observed ADS measurements at NF and SF sites. Average ammonia emission contributions to the receptor for 12 hours is expressed as

$$\left[NH_3\right]_{Lagoon,\ i,t} = \sum_{t=1}^{n}\left(\sum_{i=1}^{m}\left[NH_3\right]_{\ i,\ t}\right)/n,$$

where *i* is the number of cells and n is the number of time which contribute to receptor ammonia concentration. Thus, the averaged ammonia concentration for each contributed lagoon point source to the receptor was indicated as $[NH_3]_{Lagoon}$. The ammonia concentration at the NF site was indicated as the background concentration expressed as $[NH_3]_{NF}$. Thus, having multiple point sources in the model, the input concentration data, $[NH_3]_{NF+Lagoon}$, is defined by ammonia concentration at the NF site, $[NH_3]_{NF}$, plus the sum of concentration, $[NH_3]_{Lagoon}$, at the receptor from each contributed point sources. Therefore, the ammonia input data for estimating the

reaction rate constants between the gas-to-particle conversion process was defined as the sum of ammonia concentration from the background NF site and the emission associated with the waste lagoon.

Time Constant for Equilibrium State

The chemical composition of the sample is assumed to consist primarily of ammonium, sodium, sulfate, nitrate, and chloride, which is in equilibrium with the gaseous species. The measured gas phase concentrations can then be compared to those predicted from the chemical composition of the aerosols.[36,37,38] Estimating the time constant for an equilibrium state between gas and particle phases, we assumed that ammonia, acid gases, and fine particles are already in an equilibrium state at the NF site before reaching additional ammonia emission from a swine lagoon. After passing a swine lagoon at the experimental site, due to an additional emission of ammonia, the state between the gas and particle phases are in non-equilibrium. The thermodynamic aerosol model based on equilibrium constants has been used for predicting the concentrations of gas and particle phases at the upwind NF site as an input data for this modeling.

Gaseous ammonia in the troposphere is converted, among other processes, into ammonium aerosols by neutralization processes of ammonia with sulfuric acid, nitric acid, and hydrochloric acid in the atmosphere. In this study, we have focused on ammonium nitrate and ammonium chloride equilibrium chemical reactions (Equation 2.1, 2.2), since sulfuric acid is non-volatile at ambient conditions, and not considered. The following gas (g), liquid (l) and solid (s) phase reactions summarize ammonium aerosol production :

$$NH_3(g) + HNO_3(g) \xleftrightarrow{k_N} NH_4NO_3(s)\,or(l) \tag{2.1}$$

$$NH_3(g) + HCl(g) \xleftrightarrow{k_{Cl}} NH_4Cl(s)\,or(l) \tag{2.2}$$

$$\frac{d[HNO_3]}{dt} = -k_N \cdot [NH_3][HNO_3] : \frac{d[HNO_3]}{dt} = -k_N \cdot [NH_3][HNO_3] \tag{2.3a}$$

$$\frac{d[NH_3]}{dt} = \frac{d[HNO_3]}{dt} \tag{2.3b}$$

$$\int_{[NH_3]_{NF+Lagoon}}^{[NH_3]_{Equilibrium}} d[NH_3] = \int_{[HNO_3]_{NF}}^{[HNO_3]_{Equilibrium}} d[HNO_3] \tag{2.3c}$$

Where $[NH_3]_{NF+Lagoon}$ is the concentration of ammonia associated with the background value (i.e., $[NH_3]_{NF}$) plus ammonia emitted from the waste lagoon. $[NH_3]_T$ is the predicted equilibrium concentration of ammonia from EQUISOLV II, and

A = $[NH_3]_{NF+Lagoon}$ - $[HNO_3]_{NF}$ (unit : $\mu g/m^3$)

(Assumption:

$[NH_3]_T$-$[NH_3]_{NF+Lagoon}$ = $[HNO_3]_T$-$[HNO_3]_{NF}$).

$$\frac{d[NH_3]}{[NH_3][HNO_3]} = -k_N \cdot dt \tag{2.4a}$$

$$\int_{[NH_3]_{NF+Lagoon}}^{[NH_3]_{Equilibrium}} \frac{1}{[NH_3]_T[HNO_3]_T} d[NH_3] = -\int_o^T k_N dt \tag{2.4b}$$

$$[HNO_3]_T = [NH_3]_T - ([NH_3]_{NF+Lagoon} - [HNO_3]_{NF}) = [NH_3]_T - A \tag{2.4c}$$

$$\int_{[NH_3]_{NF+Lagoon}}^{[NH_3]_{Equilibrium}} \frac{1}{[NH_3]_T\{[NH_3]_T - A\}} d[NH_3] = -\int_o^T k_N dt \tag{2.4d}$$

Using the following boundary conditions, k_N is the rate constant of ammonium nitrate aerosol (1.83×10^{-4} $m^3/\mu mole/sec$), k_{Cl} is the rate constant of ammonium chloride aerosol (2.61×10^{-4} $m^3/\mu mole/sec$) from Baek and Aneja (2001)[23],

t=0: $\left[NH_3\right]_o = \left[NH_3\right]_{NF+Lagoon}$;

t=T : $\left[NH_3\right]_T = \left[NH_3\right]_{Equilibrium}$

(T is the equilibrium time constant from source to receptors), Thus, the equilibrium time constant, T_{H4NO3}, between NH_3 and HNO_3 aerosol is defined as :

$$T_{NH_4NO_3} = \frac{1}{A\,k_N}\ln\left[\frac{\left[NH_3\right]_{Equilibrium}\left(\left[NH_3\right]_{NF+Lagoon}-A\right)}{\left[NH_3\right]_{NF+Lagoon}\left(\left[NH_3\right]_{Equilibrium}-A\right)}\right] \quad (3)$$

Similarly, the equilibrium time constant, t_{NH4Cl}, between ammonia and nitric acid aerosol is also derived ($B = [NH_3]_{NF+Lagoon} - [HCl]_{NF}$) :

$$T_{NH_4Cl} = \frac{1}{B\,k_{Cl}}\ln\left[\frac{\left[NH_3\right]_{Equilibrium}\left(\left[NH_3\right]_{NF+Lagoon}-B\right)}{\left[NH_3\right]_{NF+Lagoon}\left(\left[NH_3\right]_{Equilibrium}-B\right)}\right] \quad (4)$$

Transfer between the gas and particle phase results in a change in the ambient concentrations and the particle phase concentration in the particles. Change in particle surface concentrations due to changes in aerosol mass of inorganic compounds in the particle also affects the equilibrium time constant. Thus, the transfer time scale between the gas and particle phases is also dependent on the ambient temperature, relative humidity, and their gas and particle phase concentrations.

Results and Discussion

Based on the kinetic rate constant between NH_3 and HNO_3 aerosols, and NH_3 and HCl aerosols, the equilibrium time constant will be estimated from the measured ammonia, acid gases, and fine particles at a commercial hog farm in the eastern North Carolina. The estimated equilibrium time constant between NH_3 and HNO_3 aerosol, and NH_3 and HCl aerosol may range from a few minutes to over a few hours under certain conditions. Even though the volatile inorganic compounds are not in equilibrium between the gas and particle phases in the real atmosphere, a thermodynamic equilibrium model, EQUISOLV II, is used in this study for predicting the equilibrium gas and particle phase concentrations. Under certain transport cases, the comparison of ammonium and nitrate concentrations with observed and predicted aerosol compositions from the thermodynamic equilibrium model will be compared.

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Particle size selective PM measurements

Heinz Fissan [1] and Thomas Kuhlbusch [2]

Abstract

Information on PM_X sources becomes more and more important in view of the recent finalized 1. Daughter Directive of the European Union related to ambient air quality. Field measurements of the new PM_{10} standard as air quality criteria for ambient particles indicate exceedances of the limit values, especially for the daily average limit value. Measurements and along with that appropriate measurement techniques are necessary a) for exposure assessments indoors, b) for emission assessments, c) for influence estimates on the local ecology, and c) for source apportionments. The presentation therefore focuses on the summary of the current state of the art of PM_X-measurement technology and will also give some information on recent and currently ongoing developments.

1 Introduction

Following the ambient air standard development in the US the European Union is discussing new ambient air standards. Daughter directives have been released which are related to limit values of sulfur dioxide, nitrogen oxides, particles and lead in ambient air (Council Directive 99/30/EC)

Whereas until 2000 ambient Total Suspended Particle (TSP) concentrations were allowed up to 150µg/m^3 as annual average and up to 300µg/m^3 with maximal 7 exceeding values as daily average, starting 2005 the new PM10 standards ask for an annual average of smaller than 40 µg/m^3 and an PM10 daily average of smaller that 50µg/m^3 with max. 35 exceeding values. Standards for PM2.5 are in preparation.

These new standards are based on the experience that larger particles are less important with respect to health effects. They are defined by collection efficiency curves with 10µm, 2.5µm and even 1µm cut off, respectively. Whereas the 2.5µm cut off always includes part of the coarse mode in ambient air, the 1µm cut off assures that only particles of the fine mode are collected (see figure 1). Some health effect experts discuss that the mass concentration in not an appropriate measure at all. They ask for measures, which are related to particle surface or even particle number.

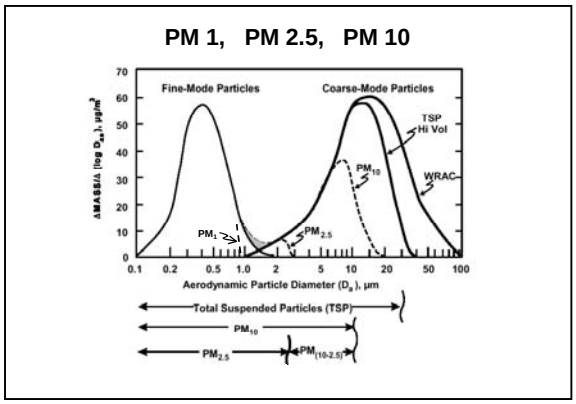

Figure 1:
Mass size distribution of ambient air and definition of PMx-standards

Although the discussed standards are referring to ambient air they will start a discussion about similar

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standards at working places and for emission control. In agriculture particulate matter is emitted in many different ways. It influences the agricultural working places as well as ambient air.

In this presentation we report about the state of the art with respect to the instrumentation for measuring the interesting quantities, but putting emphasis on more recent developments which try to solve some of the existing drawbacks of used instrumentation. Of course we will especially describe some of the new developments in our lab, which deal with reducing artifacts during particle collection, development and use of a cascade impactor especially designed for PMx-measurements in ducted emission. We will also briefly discuss the possibilities for measuring number concentrations and the corresponding size distributions. Although these latter techniques are not yet standard, they provide detailed information which helps to understand the physical formation, changes and behavior of aerosols. This is needed if appropriate measures have to be taken to improve processes leading to particle emission reductions.

2 PMx-Measurement equipment

The first step in measuring PMx-mass concentrations is always the separation of the desired PMx-size fraction. Particle separation in the size fractionating inlet is based on the inertia of particles. Separators mainly used are impactors, and sometimes cyclones. These separators should be in accordance to CEN or CFR standards (CFR 40, Part 50). The inlet to the separator for ambient air measurements has to sample the ambient particles without influence to the particle size distribution (up to particles larger than the largest particle size of the collection efficiency curve of the separator) independent of wind speed and wind direction. Since large impacting particles show the tendency to bounce, due to their high kinetic energy, it is advantageous to include PM10 and may be PM2.5 separators, in the set-up before measuring smaller PMx-fractions (e.g. PM1). We recently extended the possibilities of a commercially available High Volume Sampler by adding a PM1 stage in line with an already existing stage (John, 2002).

The particle fraction passing through the separators has to be separated from the gas phase. This is done mainly by filtration. The filters are weighted before and after a certain aerosol volume has passed through the filter. With the known flow rate, sampling time and collected particle mass the mass concentration can easily be determined. The sample can also be used to perform chemical analysis.

In a 1998 measurement campaign we sampled PM10 and PM2.5 fractions at a rural site in Spellen. The average PM_{10} and $PM_{2.5}$ mass concentration for a period of three weeks are given in figure 2a and 2b.

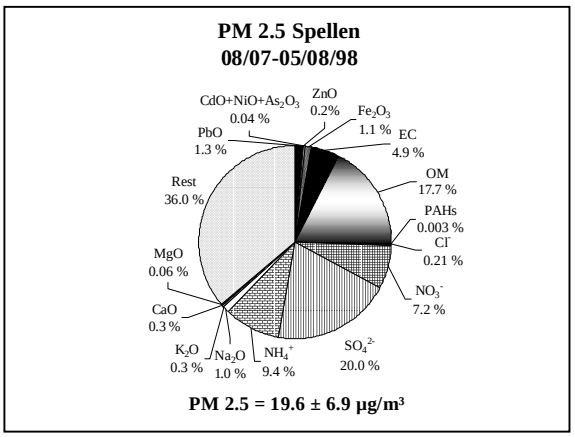

Figure 2a:

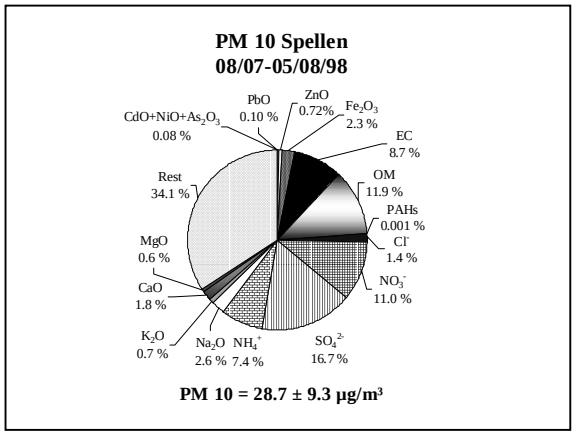

Figure 2b:
PMx-concentrations and chemical composition at a rural site

PM_{10} limit values were not exceeded during this period assuming that the PM_{10} value is representative for the annual average. Significant exceedances of the limit value for daily averages (50 µg/m³) were determined during a measurement campaign the year before (exceedances 8 of 21 days, not shown). The exceedances of values up to 140 µg/m³ were mainly due to NO_3^-, SO_4^{2-} and NH_4^+ representing 42%, 21%, and 18% of the $PM_{2.5}$ mass, respectively (Kuhlbusch et al., 2000). The average chemical composition of PM_{10} and $PM_{2.5}$ in 1998 are also shown in figure 1a and b. The main sources of NO_3^- and SO_4^{2-} are industrial, domestic, and traffic related combustion sources whereas the main source of NH_4^+ are agriculture and animal farming. When interpreting the above results it has to be noted that significant amount of particulate NH_4NO_3 may have not been sampled due to volatilization during sampling. NH_4NO_3, NH_4Cl, some organic compounds and other

semivolatile substances are always problematic to be sampled with manual or automatic filtration samplers.

Filters are always exposed to changes in temperature and relative humidity during sampling, causing artifacts which can not be controlled. In case of manual handling of the filters one tries to minimize these effects by equilibrating the filter under standard conditions before weighing. Artifact formation is especially a problem with automates, which allow quasi-on-line weighing. The mostly used instruments are β-gauge and the Tapered Element Oscillating-Microbalance (TEOM).

Humidity changes effect the sensor as well as the sampled particles. Since the adjustment to new conditions takes time memory effects may occur. Sensors of automatic devices are normally operated at elevated temperatures (e.g. 50°C) to reduce humidity effects. This measure reduces the relative humidity below the point of deliquescence in most cases reducing the influence of humidity on particle mass determination. But increasing the temperature causes semi-volatile particles to evaporate. This process has been studied and modeled in detail under defined laboratory conditions (Furuuchi et al., 2001). Measurements in the field show a mass reduction due to evaporation up to 30% compared to manual samplers.

To solve this problem we developed a concept shown in figure 3.

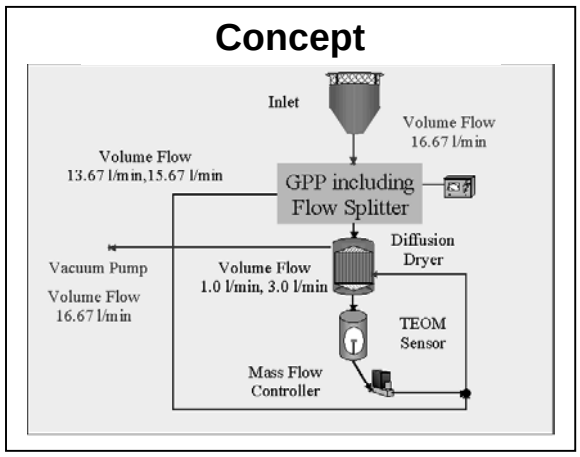

Figure 3:
Concept for measuring PMx-concentrations without artifacts

The PMx-separator is followed by a kind of chopper, which allows to take out all particles during a given time period. It is a specially designed electrostatic precipitator, which acts also as a flow splitter. The sample flow of only 1 l/min (or 3 l/min) is dried in a diffusion dryer, whereas the remaining flow is used to carry away the extracted water. The dried air is used to check the zero point of the TEOM, which is changing with the particle load. This reference measurement is used to correct the measured particle mass concentration, when the chopper is not operating.

This procedure allows the measurement of the correct particle mass concentration, if the added components do not change the thermodynamic state and chemical composition of the aerosol. Right now we are checking the performance of the single components under defined laboratory conditions.

In order to improve air quality in the environment and at working places particle emitters have to be characterized. This can relatively easily be done for point sources.

Previously, stack emissions have been monitored by measuring TSP using e.g. plane filter devices according to the VDI guideline 2066 part 7 (1993). For the determination of particle mass size distributions, emission measurements with cascade impactors or cascade cyclones have been carried out (VDI 2066 part 5, 1994).

Changes in emission reduction technology have decreased the emission of TSP, but the emission of particles smaller than 10 µm can still be above the acceptable limit. Commercially available cascade impactors or cyclones are of limited suitability for the determination of PM10 and PM2.5. They require the analysis of multiple stages and require long sampling times, due to particle distribution over many stages and volume flows of only about 1 m³/h. More than 24 hours are often necessary to obtain enough material for gravimetric analysis. Furthermore, the size cuts of the different stages are not designed to sample the exact size fractions of PM10 and PM2.5.

For more precise measurements of PMx, a new PM10/PM2.5 cascade impactor (GMU - Impaktor *johnas*) for in-stack measurements was designed and constructed (see figure 4).

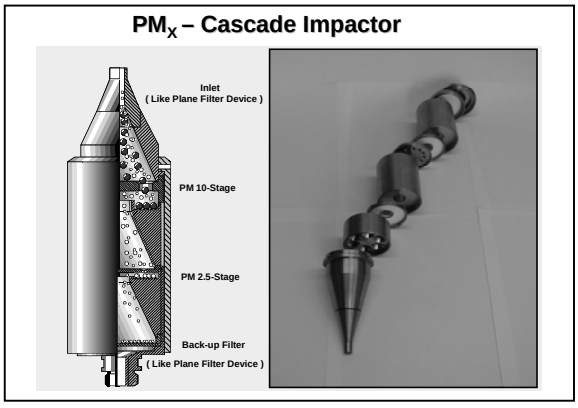

Figure 4:
PMx-Cascade impactor for point emission measurements

The PM10 and PM2.5 impactor stages were calibrated with monodisperse polystyrene latex particles (microparticles, Berlin; Duke Scientific, Palo Alto, California, USA), using optical particle counters (PCS 2000, Fa. Palas, Karlsruhe; Lasair Model 101, Particle Measuring Systems Inc., Boulder Colorado, USA). Details of the calibration process are described elsewhere (John et al., 1999).

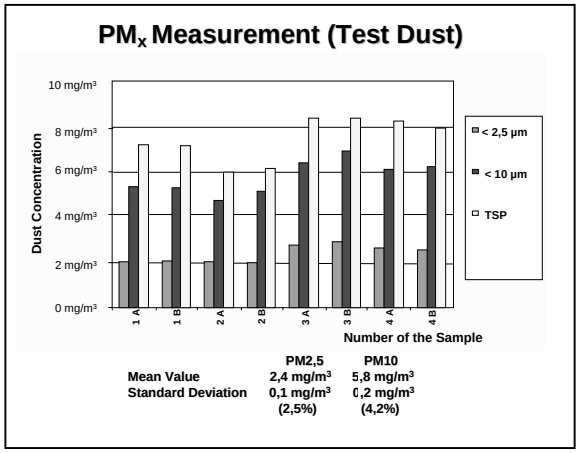

Figure 5:
Comparison of two instruments A and B site by site

Figure 5, shows the comparison of two instruments A and B challenged with test dust. The reproducibility of the instruments is very good. The advantages of the new cascade impactor are:

- It allows the direct measurement of PM10 and PM2.5 like proposed for ambient air standards.
- Relative high flow rate (3.2m^3/h) which reduces the sampling time for reduced emissions.
- The set up is similar to the plane filter device, which allows the use of the same additional equipment.
- Bounce-off and blow-off effects are minimized by collecting particles in cups. This also improves the handling of the sample.
- At the stages the flow is directed towards the centre thus avoiding particle losses at the walls.
- The possible change of cut off due to changes in temperature, pressure and gas composition is taken care of by adjusting the flow rates.

Particle number concentration

The gas to particle conversion process plays an important role especially in agriculture since emissions of NH_3 allow the formation of ammonium sulfate, - nitrate, and -chloride. To investigate these processes measurement instruments are needed which are very sensitive with respect to small particles. They have to measure number concentration, because it is much more sensitive than mass concentration. The instrument mostly used for particle number concentration measurements is the Condensation Nuclei Counter (CNC). For process investigation it is important to get information about the particle number size distribution. In the submicron size range, especially if very small particles are of interest, an important technique is the differential mobility analysis (DMA) used in the Differential Mobility Particle Sizer (DMPS) (Fissan et al., 1983).

Sizing instruments cover always only 1 ÷ 1.5 decades of particle size. In gas to particle conversion processes the particle size of interest ranges from 1nm to 1µm over three decades. Therefore a prototype of a new measurement system based on differential mobility analysis has been developed. It mainly consists of the following components:

- A new diffusion charger which improves the charge level of the smaller particles to be measured
- A radial DMA with two inlet slits which allows to extend the size range for particle size fraction for 1nm ÷ 1µm.
- Followed by an electrometer for number concentration measurement, which is easier to use in the field.

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New portable environmental fine dust analyzer for simultaneous monitoring of PM-10, PM 2.5 and PM-1

Hans Grimm [1]

Introduction

The GRIMM Model 107 is an extremely small and portable particle analyzer. The Model 107 is specifically designed for PM-10, PM-2.5 and PM-1 environmental ambient air analysis. It performs particulate measurements using 90° laser light scattering technology. Using this type of technology enables the Model 107 to make very precise “cut points” for all three PM size classifications. This unique patented system allows the user to collect all three PM fractions simultaneously without changing sampling heads or weighing filters. However, the Model 107 is the only PM monitor to offer dual technology consisting of both optical and gravimetrical analysis. The Model 107 incorporates a removable 47 mm PTFE filter which allows the user to verify the optical analysis gravimetrically, as well as, providing the option for other chemical analysis on the collected residue.

Present method

Most classical methods collect aerosols at a given volume on a filter and the dust mass is identified as "MASS" in microgram/m³. This (mostly) high volume sampler technology, however, limits oneself to the pre-selected sampling head, the specific cut point and the surrounding wind speed. Greater wind and / or humidity variations, more secondary and/or anthropogenic particulate and other parameters (topology, acid aerosols) can also change the results considerably. In addition, the performance decreases with the reduction of the particle size cut point of the sampling head, since the remaining mass of very small particulate continues to decreases and therefore harder to weigh.

New technology

A new technology permits real-time dust monitoring of particles from 0,3 microns to 20 µm in many different size channels. The particle size of the aerosol is determined by light scattering, this means the use of a laser, collector lenses and mirrors, a photo diode and an attached 15-channel pulse height analyzer. These results are classified as "counts" and expressed as counts/liter. This optical bench is manufactured in such a way that it is unaffected by shock and vibration.

From this count distribution all other information, such as the TSP (or inhalable), the PM-10 (or thoracic) and the PM-2.5 (or alveolic / respirable) fraction are obtained. Even the proposed PM-1 (submicron fraction) can be determined and constantly displayed.

Continuous air is drawn into the monitor by a volume-controlled pump. All particulates in this air pass the above described sample cell and then are captured by a removable PTFE Filter. This filter permits a later chemical and/or particle analysis, but also the gravimetrical correlation to the "local" aerosol conditions. With this unique technique a standard instrument can be adapted to many different aerosol configurations.

An unique “sheath air” system not only keeps the optical surface of the measuring cell clean, it also helps to reduce high sample air humidity and is used as AUTOZERO for the optical drift control.

The complete system with battery is packed is such a way, that mobility is assured. This small portable analyzer is only 9.5 x 4.7 x2.5 in and with battery only 5 lb.

All results obtained as well as the instrument performance are exported, via interface, to an external computer. Not only can data be shown in real time in WINDOW based Software with these fine dust monitors in counts or mass of all 15-size channels, but also all Environmental mass values can be shown in numeric and graphic mode.

To assure optimal results, temperature, humidity and wind-speed sensors can be attached to the monitor and used for background information. All

these values, as well as the internal behavior of the unit, are shown each minute on the PC.

[1] Grimm Aerosol Technik GmbH & Co. KG, Dorfstraße 9, D-83404 Ainring, Germany

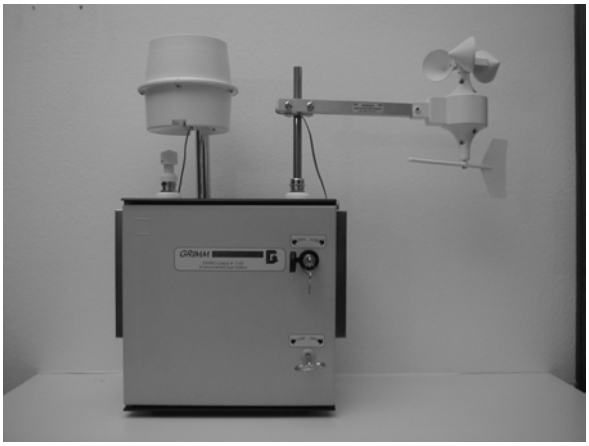

Figure 1:
This photo shows the 107 system in the weather housing 165 with sensors for precipitation on the left, and for wind speed / wind direction on the right

System configuration

- The complete 107 system for outdoor use (figure 3 + 4) is consisting of the 107 PM dust monitor, the 165 fiberglass housing, the drying temperature control system, sensors for humidity and temperature, and the 170M sampling head. Displays real time data of PM-10 and PM-2.5 as quickly as every six seconds.
- Auto-zeros and performs system self-diagnostics at the start of each analysis.
- Removable EPA approved PM-2.5 PTFE filter (diameter: 47 mm)
- Removable Data cards, can hold up to one year of data.

Figure 2:
107 system installed in a shipping harbour

Figure 3:
107 system is used at a truck stop control point

Benefits and features

- Displays real time data of PM-10 and PM-2.5 as quickly as every six seconds.
- Portable, the monitoring units weighs only 2.4 kg and is removable from the weather housing for quick scanning jobs, or indoor monitoring.
- Remote control via tel-modem
- Remote instrument system analysis
- Expandable for Climatic accessories.

Results

Obtained results from many national and international organizations are existing and these data were compared with existing REFERENCE INSTRUMENTS.

Below are PM10 comparison results (figure 4) obtained in Europe in four 2000. The reference system here was a Swiss very high volume sampler, 3 different locations during this one year were selected.

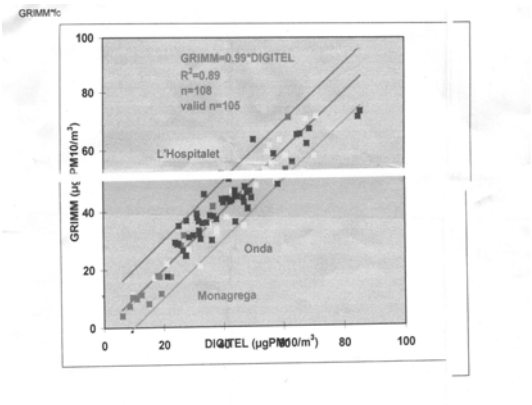

Figure 4:
Comparison results

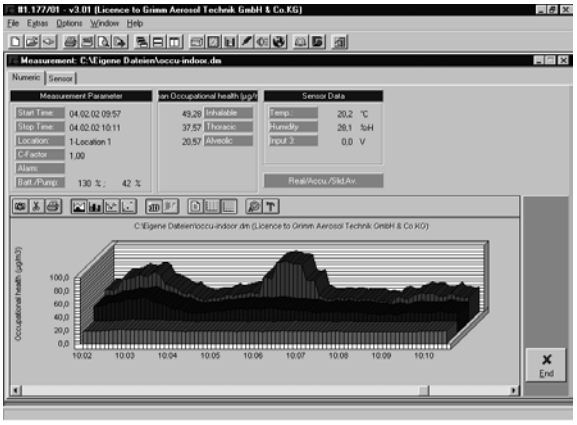

Figure 5:
The new 32-bit version Network software shows the short term result of 10 min.

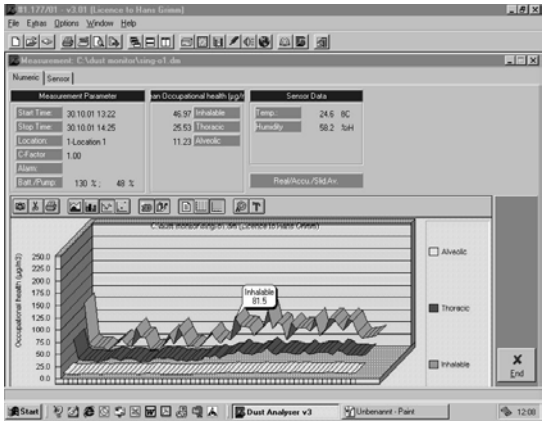

Figure 6
Environmental results for respirable, thoracic and alveolic particles

Real time portable aerosol particle sizer (counter) from 0,005 to 20 µm

Uwe Golz [1]

Introduction

This new portable real time PARTICLE COUNTER SYSTEM is designed for mobility and easy field use. Its integrated battery assures hours of uninterrupted operation. The obtained results are stored on the removable data logger card, permitting an easy data transfer to any PC by means of the user friendly powerful software that allows easy operation.

This system is based on the combination of two technologies of particle counters: An **Ultrafine Particle Counter** and a **Fine Particle Counter** are joined together to one system by means of the software.

System

The new measuring system consists of a Laser Aerosol Spectrometer, model 1.108, and an Ultrafine Particle Counter, model 5.400:

The Aerosol Spectrometer takes a defined air sample by a volumetrically controlled pump and measures the airborne particles continuously by scattered light in its laser measuring cell. The measuring range of the particle sizes is between 0,3…>20 µm in 15 size channels. The Ultrafine Particle Counter measures as Condensation Nucleus Counter in a range between 5…350 nm. Together with a DMA, model 5.500, it is possible now to measure also the size channels in the nanometer range (System SMPS+C). A personal computer synchronizes both measuring systems and records the measured results. The software delivers the measured results in particle number per size channel for the complete measuring range from 0,005 to 20 µm.

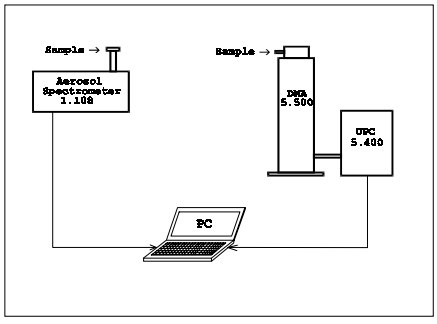

Figure 1:
Scheme of the wide range measuring system

System Configuration

Instead of using several individual components combining to an operational system, all necessary items and appropriate functions have been combined into one portable system consisting of:

A) 1.108 Portable Aerosol Spectrometer:

The enhanced Aerosol Spectrometers are the world's smallest family of battery operated portable fine dust monitors available on the today's market. This unit measures highest particle concentrations up to 2.000.000 particles/liter as well as a single particle. The results are displayed as counts (p/l) or as mass distribution up to 100.000 µg/m^3 in 15 different size channels.

Range: 1 to 2.000.000 particle/liter of >0,3…>20 µm in following channels:
(0,3/0,4/0,5/0,65/0,8/1,0/1,6/2,0/3,05,0/7,5/10,0/15,0/20,0 µm)

The model 1.108 uses a light-scattering technology for single particle counts, whereby a semiconductor-laser serves as the light-source. The scattering signal from the particle passing through the laser beam is collected at 90° by a mirror and transferred to a recipient diode. The signal of the diode passes, after a corresponding reinforcement, a multi-channel size classifier. A pulse height analyser then classifies the signal transmitted in each channel. These counts can be displayed and are also stored in the data storage card and may be transferred via RS 232 for further analysis.

The ambient air, to be analysed, is drawn into the unit via an internal volume controlled pump at a rate of 1,2 l/min. The sample passes through the sample cell, past the laser diode detector and is collected into a 47 mm Teflon filter. The entire sample is collected on this PTFE-filter, which can be analysed gravimetrically afterwards for verification of the reported aerosol mass. Additionally, further chemical analysis can be performed on the deposited residue. The pump also generates the necessary clean sheath air, which is filtered and passes through the sheath air regulator back into the optical chamber. This is to ensure that no dust contamination gets in contact with the laser optical assembly. This particle free airflow is also for the reference zero test during the auto calibration.

[1] Grimm Aerosol Technik GmbH & Co. KG, Dorfstraße 9, D-83404 Ainring

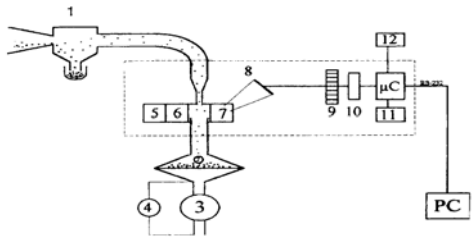

Figure 2:
The diagram below shows the concept of a typical FINE DUST MONITOR:

(1) Sample inlet
(2) Teflon filter
(3) Constant flow pump
(4) Sheath air
(5)(6)(7) Optical bench (laser diode, lenses, light trap)
(8) Photo diode
(9) Pulse height analyser
(10) Size range classifier
(11) Data storage card Display

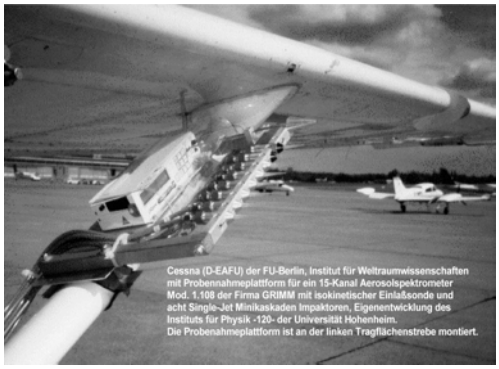

Figure3:
This photo shows a 1.108 and 8 step impactor attached to an airplane wing for "environmental" control (University Hohenheim, D-Stuttgart)

Software Presentation of the 1.108 Aerosol Spectrometer:

We have developed our own 32-bit software to manage the GRIMM Aerosol Spectrometer. The minimum requirement is a Pentium II Processor and MS-WIN 9x. All set-up phases are supported in our HELP and the TUTORIAL guide. Generally speaking, the program enables the user to set all operating parameters via specific driver logic such as:

- User defined operating modes on the monitor, manual or automated starting (even remotely), pre-selectable alarm levels, sampling location, optional sensor setting, to include as well the data display mode in the count or mass mode.
- The start/stop pre-selections and measurement intervals can be defined from 6 s to 60 min (optional 1 s intervals). Even the service data are stored to be viewed at a later date. There is also a full displayable protocol via PC of the entire performance (available at any time).
- The control of the monitor performance is fully automated with a zero count test at the start. This is to include the sample air flow control, the filter presence and particle size range as well. There is even a battery power management and battery recharge logic.

Data Presentation Possibilities via PC:

- Simultaneous particle size distributions in count modes of 15 channels,
- Mass distribution in accumulative and differential mode (µg/m^3) in 15 channels,
- Simultaneous readings of Inhalable, Thoracic and Alveolic values in real time,
- Simultaneous readings of EPA environmental PM_{10} and $PM_{2,5}$ in real time,
- Mean values,
- Calculated surface area of particles in m^2/m^3,
- Optional environmental sensors (Temperature and humidity, wind speed).

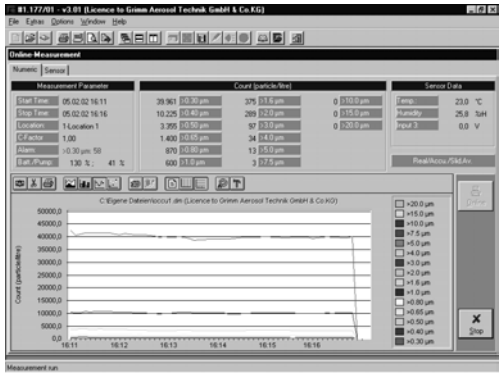

Figure 4:
Example of 5 min. monitoring data in the COUNT mode

B) 5.400/5.500 Scanning Mobility Particle Sizer+C:

Introduction

This new family of portable real time SEQUENTIAL MOBILITY PARTICLE COUNTER and SIZER (SMPS+C) is designed for mobility and easy field use. An integrated battery assures hours of operation, a data logger system storage of all obtained results and a user friendly powerful software easy

operation. This technology not only simplifies the SMPS operation, but it permits new on site application monitoring up to a remote wireless telephone operation.

Instrument

Instead of using several individual components combining to an operational system, all necessary items and appropriate functions have been combined into one portable system. This means the well known principle of "Nucleus Condensation Counting" (Willeke and Baron, 1993) was improved in such a way that:

1. The alcohol saturation chamber gets only as much moisture as needed, eliminating possible overspill and consequently contamination of the condenser and the optic,
2. The alcohol tank is integrated into the system, the level and consumption controlled and an automatic refill system assures long term operation,
3. The sample outlet air has an alcohol odor adsorber,
4. A new moisture elimination system assures long reliable field measurements,
5. An internal heat exchange system reduces drastically the electric power consumption, permitting hours of continuous battery operation,
6. An integrated air and power supply control system permits the attachment of any of our classifiers, reducing not only the overall size, but also handling and system manipulation,
7. An additional port permits the attachment of external gas and/or climatic sensors, helping a better judgement of the sampling and measurement data obtained,
8. An integrated microprocessor system permits the operation of the complete system without an attached computer, status lights and a digital display show the measurement conditions,
9. Measurement, instrument and sensor data of up to ½ year can be stored on a removable data storage card,
10. The single counter and even the complete SMPS+C system can be operated remotely via a telephone modem,
11. A powerful 32 bit software makes data acquisition and analysis (including well-known corrections, e.g. Wiedensohler and Fissan, 1988) very practicable.

Table 1:
Performance of the Particle Counter

Parameter	Range
Min. particle size in [nm]	5
Max. concentration [P/l]	10^{10}
Count Rate [P/l]	10^{7}
Sample flow rate [l/min]	0.3 or 1.5

The battery driven all in one system SMPS+C (Sequential Mobility Particle Counter + Sizer) is developed and fitted with newest technology. The very low power consumption heating/cooling unit with an intelligent on board control system allows very long battery driven stand alone times. The complete control and supply of the classifier is integrated on board the unit as well. Different classifiers from 5 – 900 nm are available (Reischl, 1991).

With this complete mobile and portable system for the first time field test even under rough treatment got possible. The construction of the system even allows measurements during cross-country trips and flights in aeroplanes. The measured data and the data of external sensors are stored in the memory of the internal microcomputer. For longer measurements a removable data storage card could be connected to the system via pcmcia-socket. So the system could be driven as stand alone unit even without a connection to a computer. Data and operation commands could be send and received via an optional customary mobile modem pc-card. The SMPS could be completed to a remote operated field measurement system.

Figure 5:
Portable Sequential Mobility Particle Counter with small Classifier, external sensor and mobile phone adapter.

Software Presentation of SMPS+C:

This program operates all GRIMM ultra-fine particle counters and SMPS+C systems. The minimal requirement is a standard Pentium II processor and WINDOW's 98.

- Control of the UPC

The counter system with sample air flow and interval, date and time, location ID, power management, battery charging, automatic zero-test and a selectable alarm. The selection of any auto start and stop function as well as any measurement interval times of 1 to 15 min can be predefined via computer.

- Control of the SMPS+C system

First the sheet air supply and zero system tests are made, then the desired channels and selectable measurement range size can be set in accordance to the attached classifier and entered in. Then you enter in the amount scans and time range from one channel to the next, or select the auto-mode.

- Data presentation possibilities such as
  - Mean values of all spectras obtained
  - Numeric and graphic data presentation in the count mode (up to 255 different size channels)
  - Immediate count presentation at a specific size
  - Data presentation with transformation correction
  - Printable results via RS 232
  - Stored data download of data card
  - Data exportation in ASCII code

- Conversion calculations of count results obtained from any data file for:
  - Multiple charge calculations
  - Auto size channel for transformation calculations
  - Log-normal size distribution calculations

Examples of results:

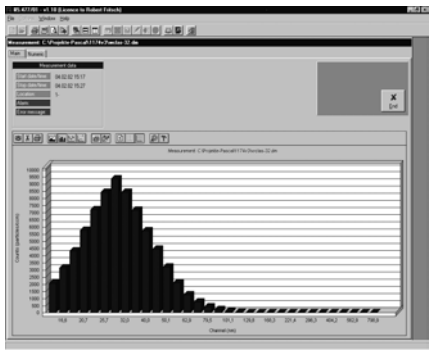

Figure 6:
The image below displays a transfer conversion of 15 nm monodisperse NaCl Concentration

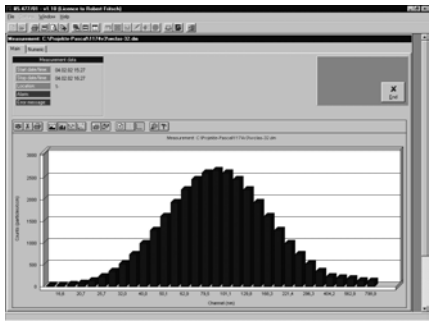

Figure 7:
The image below shows a quick scan of a 32 point size distribution

Conclusions

The upper measurable particle size of any SMPS+C system is limited to sub-micron particle range. With the Aerosol Spectrometer and the SMPS+C in one system a new generation of field and laboratory particle counter and sizer was built up focused on mobility, ruggedness, stand alone operation and up-to-date compact high-tech. The application ranges from climate research, workplace and medical studies to emission investigations such as exhaust gas and filter-efficiency tests.

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Wiedensohler, A., and Fissan, H., (1988): Aerosol Charging in High Purity Gases. J. Aerosol Sci. 19, 867-870.

Reischl, G. P. (1991): Measurement of Ambient Aerosols by the Differential Mobility Analyzer Method: Concepts and Realization Criteria for the Size Range Between 2 and 500 nm. Aerosol Sci. Tech. 14, 5-24.

Evaluation of a new method for personal time and size resolved dust measurements at workplaces

W. Koch, W. Dunkhorst, H. Lödding [1]

Abstract

Three particle size fractions of the airborne dust are defined in European and US standards for health-related dust measurements at the workplace: the respirable, the thoracic, and the inhalable fraction. In 1999 we introduced a novel instrument for personal, time-resolved concentration monitoring and sampling of these three fractions. The instrument combines inertial classification, filter sampling and photometric aerosol detection. It consists of a two-stage virtual impactor (cut-off diameters of 4 and 10 µm), three filters, and three light scattering photometers. The filter material can be chosen in order to meet the requirements of the respective analytical method: gravimetry, chemical, or microbiological analysis. In this paper we describe the instrument and we present recent data on calibration and validation tests as well as results of field measurements at real workplaces. The instrument fulfills the requirements of the European standards directly when operated as an area monitor. When the instrument is used as a personal monitor a constant correction factor has to be applied to the extrathoracic fraction of the dust collected by the sampler in order to achieve compatibility with the health related sampling standards. Examples from various field studies show that a large body of information is available from the measurements with this new instrument.

Keywords: Dust measurement, particle size fraction, filter, time resolution, aerosol photometer, personal sampling

Zusammenfassung

In europäischen und amerikanischen Standards sind drei Staubfraktionen zur wirkungsbezogenen Messung von Stäuben und Aerosolen am Arbeitsplatz definiert. Diese sind: die alveolengängige, die thoraxgängige und die einatembare Fraktion. Die Arbeit beschreibt ein neues Meßververfahren, mit dem diese drei Fraktionen des luftgetragenen Staubes gesammelt und nach gängigen gravimetrischen, chemischen oder mikrobiologischen Analyse-verfahren ausgewertet werden können, gleichzeitig aber auch zeitlich aufgelöst gemessen werden. Das in 1999 publizierte Verfahren ist eine Kombination aus Inertialklassierung und Konzentrationsanreicherung mittels virtuellem Impaktor, Filtersammlung und Aerosolphotometrie. Es besteht aus einem 2-stufigen virtuellen Impaktor, drei Filtern und drei Streulichtphotometern. Das Gerät kann als personengetragenes und stationäres Messgerät betrieben werden. In dieser Arbeit wird das Gerät kurz beschrieben, und es werden die Ergebnisse von neueren Kalibrier- und Testmessungen sowie Messungen an verschiedenen (landwirtschaftlichen) Arbeitsplätzen vorgestellt. Das Gerätes erfüllt die Anforderungen der Europäischen Norm unmittelbar, wenn es als stationäres Gerät betrieben wird. Bei Einsatz als personengetragenes Gerät ist für die gemessene Grobfraktion des Staubes zur Erreichung der Normenkompatibiliät ein konstanter Korrekturfaktor anzuwenden. Verschiedene Einsatzbeispiele im Feld belegen den hohen Informationsgehalt, der mit den Messungen verbunden ist.

Schlüsselworte: Staubmessung, Partikelgrößenfraktionierung, Filter, Zeitauflösung, Photometer, personengetragen, Validierung

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Introduction

In the mid-1990s the International Standards Organisation (ISO), the Comité Européen de Normalisation (CEN) and the American Conference of Governmental Industrial Hygienists (ACGIH), defined three curves representing health related particle size fractions of the total suspended particulate matter. They are: (a) the *inhalable* fraction, defining the particle fraction that may enter the nose and mouth during breathing; (b) the *thoracic* fraction, defining the particle fraction that may penetrate beyond the larynx and so reach the lung; and (c) the *respirable* fraction, defining the particle fraction that may penetrate to the gas exchange region of the lung. These curves become 'target' curves by which to define the desired particle size selectivity of a sampling instrument that provides a relevant measure of exposure. The three curves, and their application in OELs and sampling instruments, are rapidly gaining acceptance for exposure assessment at workplaces. From this basic curves two additional physilogically relevant size fractions can be calculated: the tracheobronchial and the extrathoracic fraction.

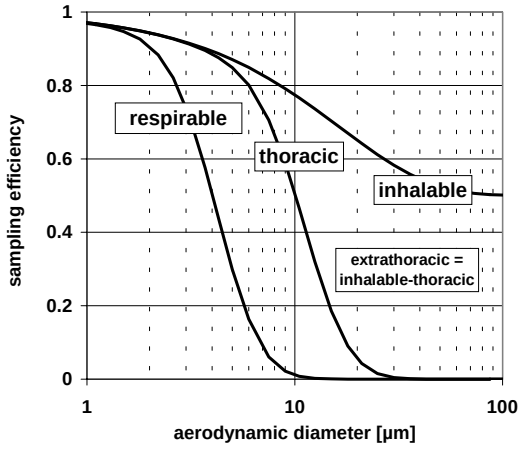

Figure 1:
Health related sampling conventions

Personal sampling as well as area sampling is applied for exposure measurements, both methods having their specif advantage. Personal sampling is recognized as the ultimate way of personal exposure assessment particularly, in the case of localized dust sources and varying tasks of the worker. Area sampling has its advantage for general characterization of working areas, the determination of long term trends or the assessment of dust removal measures taken. In general the concentrations are represented as time weighted 8 hour average values determined by evaluating the mass of the respective particle size fraction collected by the instrument. However, the ability of time resolved concentration monitoring would significantly increase the body of information usefull for characterization of the personal exposure situation, for source localization and the quick assessment of abatement strategies.

Description of the instrument

The instrumentation described in the literature does only partially fulfill the desirable properties mentioned above. Most of the instruments are pure sampling devices, though some of them are desigend to sample all three health related dust fractions. There are no (personal) monitors available enabling time and particle size resolved sampling and concentration monitoring simultaneously.

Therefore, we have designed a new instrument (Respicon), that combines inertial size classification and the standard gravimetric dust measuring procedure with the principle of optical aerosol detection. The instrument has the following properties:

- Direct aerodynamic classification of the airborne dust in three size classes: respirable, thoracic, and inhalable fraction.
- Sampling of the particle size fractions on three filters for direct gravimetric, chemical, microbiological or mircroscopical evaluation.
- Time resolved measurement of the concentration of the three dust fractions using aerosol photometry.
- In-situ calibration of the photometers via the gravimetric or chemical evaluation of the filter samples to transform the photometer signals into concentration values.
- On-site data acquisition using a portable data logger; central data evaluation on a PC.
- Suitable for area and personal monitoring.

The instrument consists of three separate components: the sensor and sampling unit, the data logger and the sampling pump (see figure 2). The sensor and sampling unit is carried close to the persons breathing zone. The data logger and the pump are worn at the girdle. The sensing unit consists of an annular slit inlet, a two-stage virtual impactor (cut-off diameters of 4.5 and 10 µm), three filters, and three constant angle light scattering photometers (wavelength 780 nm, scattering angle 90°). The concentration of the three size fractions are calculated from the masses deposited on the three filters. The virtual impactor serves as a particle size classifier and as a particle concentrator for the respective coarse fraction. This enrichment compensates for the drawback of photo-

metric measurement of coarse particles namely the decreasing particle mass-based photometric sensitivity with increasing particle diameter. The optical sensors are calibrated in-situ via the mass concentrations obtained gravimetrically from the filter samples. The device operates at a flow rate of 3.1 l/min. A detailed description of the instrument is published elsewhere (Koch et. al. 1999).

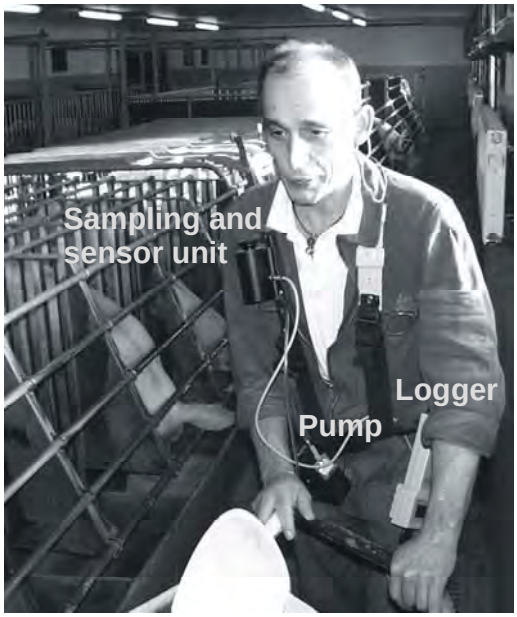

Figure 2:
The Respicon used as a personal aerosol monitor.

Data on the sampling performance

Various investigations have been performed to validate the instrument's size selective sampling performance when operated as an area sampler. In laboratory tests Koch et. al (1999) characterized the instrument under nearly calm air sampling conditions whereas Li et al. (2000) assessed the instrument in a small wind tunnel at wind velocities of 0.55 and 1 m/s. The test results presented in figure 3 suggest good compliance of the instruments sampling characteristics with the definition curves according to EN481. Due to the instrument's omnidirectional inlet the sampling performance is independent of the wind direction.

However, from the data obtained under free field sampling conditions we cannot automatically assume the same sampling performance of the instrument when operated as a personal sampler. Here, the instrument is mounted on the body forming an integral part of the sampler which can affect the motion of particles with high inertia i.e. the inhalable fraction. Also, the Respicon's omnidirectional sampling inlet may cause a reduction in sampling efficiency for very large particles due to shielding effects of the body. There exist some validation data for the inhalable fraction when the instrument was tested as a personal sampler. The data were obtained in a laboratory wind tunnel study following the test procedure standardized under prEN 13205 (1999) as well as in a field study performed in the frame of a European Research project. In both studies the Respicon was mounted on a real sized mannequin and the concentrations measured by the Respicon were compared with those obtained by reference procedures. Monodisperse test aerosols of two different particle diameters were used in the wind tunnel study. Here, the reference method was isokinetic sampling. The field study was performed in various environments characterized by aerosols of different coarsenes. The reference concentration was determined by sampling through the mouth of the mannequin at a constant flow rate of 20 l/min simulating real human breathing behavior. This is the sampling procedure by means of which the inhalability criterion was developed in the mid eighties and thus is the reference method by definition (Vincent, 1989).

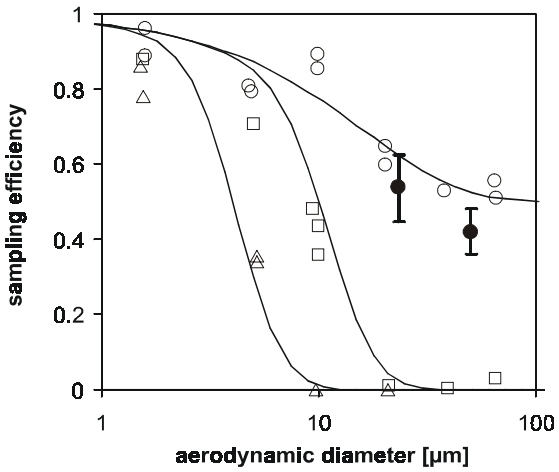

Figure 3:
Calibration and performance data obtaind in laboratory studies. Open symbols: area sampling (Li et al., 2000) ; triangles: respirable fraction, squares: thoracic fraction, circles: inhalable fraction. Full symbols: personal sampling.

The full squares in figure 3 represent the two data points of the laboratory study. These data suggest that the Respicon is undersampling the inhalable concentration when operated as a personal sampler.

The field study indicated the same trend. Linear regression analysis of the complete set of data pairs revealed the following relationship (see figure 4)

$$I_{\text{Reference}} = 1.51\ I_{\text{Respicon}}\ , \quad R^2 = 0.98$$

where I is the concentration of the inhalable fraction as measured by the Respicon, respectively the reference method. A more detailed analysis of the data showed that the ratio between the reference method and the Respicon is dependent on the fraction of extrathoracic particles (i.e. particles larger than 10 µm) present in the aerosol to be measured which is physically plausible. The concentration ratio, reference to Respicon, is one when the extrathoracic fraction is zero (i.e. the particles are all smaller than 10 µm). The ratio increases to a value of approximately two when the aerosol consists completely of extrathoracic particles, i.e. the extrathoracic fraction is one. This tendency of undersampling can be compensated for by applying an overall empirical correction factor of 1.7 to the concentration value of the extrathoracic fraction as measured by the Respicon. The transformed field data are shown as circles in figure 4. The regression results in a nearly one-to-one correspondance with the reference method:

$$I_{\text{Reference}} = 1.05 \; I_{\text{Respicon}} \;, \quad R^2 = 0.98$$

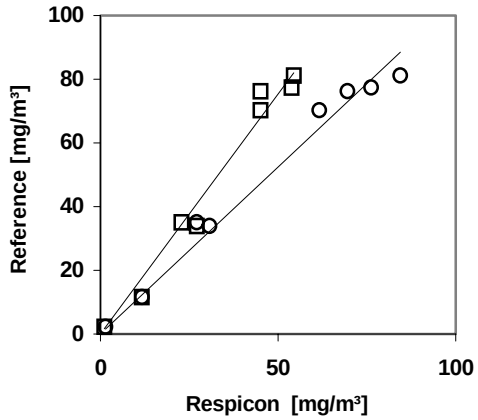

Figure 4:
Results of a field validation study for the inhalable fraction; comparison with a reference method. Squares: raw data; circles: transformed data.

Similar result were obtained by Rando (2002) in field measurements invloving wood dust. They used the IOM sampler (Instiute of Occupational Medicine) as a reference personal sampler. For the validation of the IOM sampler as an inhalable personal sampler we refer to Kenny et al. (1997). Rando obtained a regression coefficient of 1.09 for the inhalable dust when comparing the Respicon with the IOM. He applied a correction factor of 1.5 to the extrathoracic concentrations measured by the Respicon which is very consistent with our findings. In addition to the inhalable fraction Rando carried out comparisons between Respicon and reference sampler for the thoracic and the respirable fraction as well and found good agreement.

In conclusion, when operated as an area sampler the Respicon fulfills the sampling requirements of the European Standard (CEN 481, 1992) directly for all three health related size fractions. When the Respicon is used as a personal sampler the existing field validation studies suggest good performance for the respirable and the thoracic fraction. Good compliance with the inhalability criterion is achieved when a correction factor of 1.7 is applied to the extrathoracic concentration and when this corrected value is added to the thoracic concentration to obtain the inhalable concentration.

Examples of field data

The instrument has been used in many different workplace environments including those in agriculture and food industry. The plots in figure 5 show examples of the personal exposure of individuals working in animal production environments for poultry, pigs and cattle. The person have been performing routine operations such as feeding, cleaning and (partly) disinfection. For the seek of readability we have plotted only the inhalable and the respirable dust fraction as these are the two fractions for which limit values exist. The personal exposure is characterized by a high degree of temporal fluctuation. The fraction of respirable particles is generally very low. The absolute concentrations are ranked in an order as to be expected: the poultry stable showing the highest concentration, followed by the pig and the cattle stable (see Table 1).

Table 1:
Measured average personal exposure concentration in mg/m³ in different animal stables.

	Poultry	Pig	Cattle
Respirable	0.7	0.5	0.6
Thoracic	10.1	2.0	0.8
Inhalable	21.8	6.5	2.7

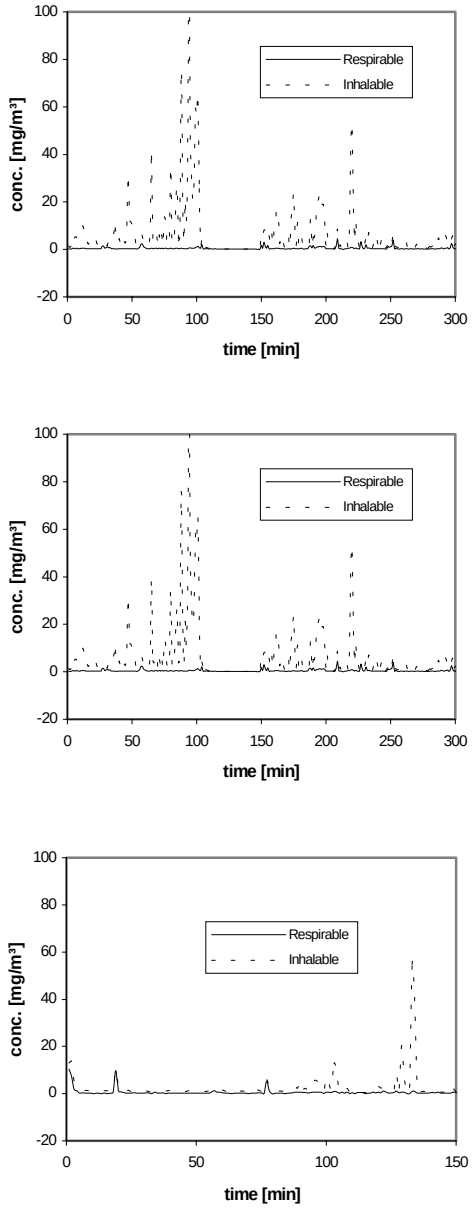

Figure 5:
Personal dust exposure in different farm environments: poultry, pig, cattle stable.

The largest differences exist for the inhalable and the thoracic fraction representing the coarse particles emitted to a large extent by the acitivities of the animals while the person is inside the stable. The con

centration of respirable particles seem to be a general background although much higher than the outdoor air concentration. There is not much correlation between the concentration of inhalable and respirable particles.

The time resolution capability of the instrument enables identification and characterization of the dust emission potential of specific operations or production processes. This is shown in figure 6 representing an example of a series of measurements carried out by Jaggy (1996). The measurements were done in the frame of animal hygiene in horse stables. One specific aim was the characterization of the exposure of horses in their boxes to dust and the correlation of the exposure to feeding and maintenance operations carried out. The concentration of the respirable fraction is generally very low. It can be seen that a significant exposure to inhalable and thoracic dust occurs after fresh straw has been put into the box. The height and the duration of the concentration peaks are quite typical and have been measured repeatedly on different days during the campaign.

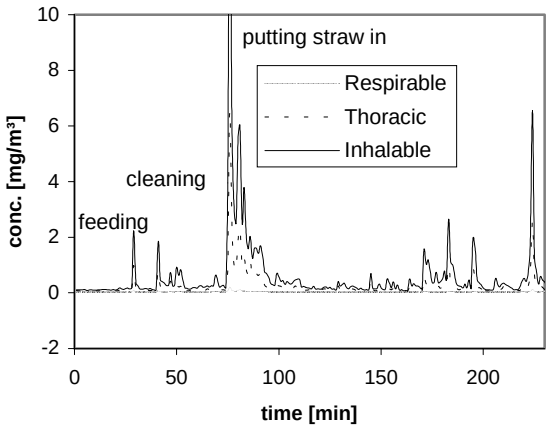

Figure 6:
Dust peaks generated by various activities in a horse box.

A third example of a field measurement demonstrates the necessity of personal sampling for proper exposure assessment. Measurements carried out in a bakery were aimed at comparing the results of area and personal monitoring, and, in addition, comparing the Respicon with dust sampling instruments used as standard instruments in Germany (VC 25, PPG; BIA). Three instruments were located at a position expected to be representative for the dust situation in the bakery: a high volume total dust sampler (VC 25), a low volume filter cassette (PPG), and a Respicon. A second Respicon was worn by a baker doing his normal job. The results showed a big difference between the personal exposure situation measured by the personal Respicon compared to what was measured by the area sampling instruments. The personal exposure shows a much more pronounced fluctuation of the concentration and the size distribution. The coarse particle concentrations are generally higher (figure 7)

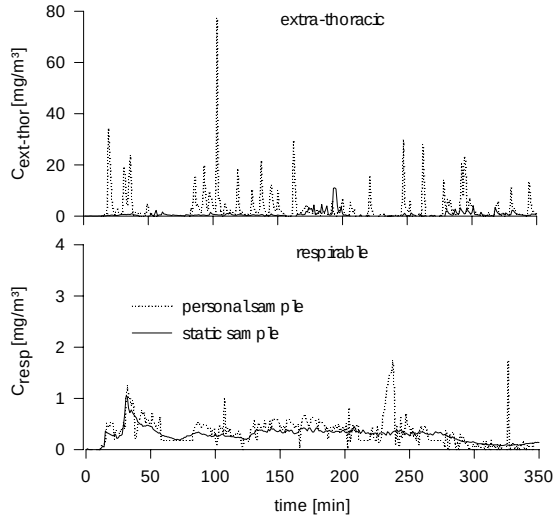

Figure 7:
Comparison of personal and area monitoring.

Table 2:
Exposure concentrations in mg/m³ measured in a bakery: personal versus area monitoring ; instrument inter-comparison.

Instrument		Size Fraction Resp.	Thor.	Inh.	Ext.-Thor.
Respicon	pers.	0.32	1.16	4.39	3.23
Respicon	area	0.28	0.74	1.85	1.12
PPG	area	-	-	1.87	-
VC25	area	-	-	1.42	-

Similar differences were obtained by comparing the shift average values determined gravimetrically from the filter samples (see Table 2). The extrathoracic fraction of the personal dust exposure was, on the average, a factor of three higher than would have been predicted from area samples. The coarse particle exposure, mainly flour dust, is determined by the individual working process. The concentrations of the respirable fraction are nearly identical, since the fine particles are spread homogeneously all over the bakery. Unlike the concentrations of the coarse fractions, the respirable concentration, as measured by the personal monitor, does not show strong fluctuations indicating a different source. Indeed, the respirable fraction consist mainly of condensed fat and baking oil particles. This example does also show the good agreement between the results of the Respicon and the German reference samplers.

Summary

In this paper we presented a laboratory and field evaluation of a versatile aerosol monitoring system combining aerodynamic size classification, filter sampling and aerosol photometry. The instrument can be used as an area and as a personal monitor. The sampling performance is in accordance with existing guidelines on health related dust measurement. The instrument operates as a sampler in combination with all kinds of off-line evaluation methods. In addition, due to its time resolution capabilities the exposure pattern can be analysed in great detail. This provides a useful data base for measures to be taken in view of reducing exposure to farm workers or improving animal hygiene. Future instrument development is aiming at a further miniaturization of the sensor head and the data logger. A reduction of the inlet flow rate to 2 l/min will also allow the use of smaller pumps.

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Determination of backscatter and fluorescence cross-sections of biological aerosols

Reiner Weichert [1]

Abstract

There is a significant interest to develop fast detection and identification capabilities for biological aerosols - like LIDAR or sampling optical sensors. Biological aerosols like pollen or bacteria can be differentiated from non-biological aerosols by fluorescence. Several organic substances in biological particles emit fluorescent light if illuminated with ultraviolet radiation. If the content of these substances in different biological particulate matter is different, then differentiation between various bacteria should be possible.

An instrument has been developed, which is capable to generate aerosols of single bacteria in concentrations of some 10^{10} cells/m^3, to illuminate an optically defined measuring volume of this aerosol with continuous ultraviolet light from a Xenon lamp - the wavelength being adjustable by a monochromator between 220 and 570 nm - and to detect the emitted fluorescence spectra in the UV and visible light. In addition, backscatter of the aerosol at 1064 nm laser-wavelength is measured giving additional information about the bacteria. The concentration of the aerosol is determined by sampling on nuclepore membranes and counting the particles in a SEM. To obtain absolute fluorescence cross sections of the bacteria, the instrument had to be calibrated. Some details of this wearisome procedure will be illustrated.

1 Introduction

Fluorescence of biological aerosols had been attributed to the three amino acids tyrosine, tryptophane and phenylalanine [1], but more than a dozen of different substances are now identified contributing to bio-fluorescence [2]. The excitation wavelengths (most in the UV) and emission wavelengths (UV and visible light) of the different substances are different, therefore the fluorescence spectra of biological aerosols are expected to change with changing excitation wavelength and with changing composition of the biological aerosols. Experimental results are contradictory: Bronk et al. [3] found a characteristic difference between the fluorescence spectra of the bacteria b. subtilis and e. coli, both at excitation wavelength of 280 nm, but Cheng et al. [4], using nearly the same excitation wavelength of 275 nm, obtained identical spectra for b. subtilis and e. coli. In the two papers, however, single particles and pulsed lasers have been used, i.e. the energy input into the bacteria was so high that photodegradation of the bacteria [5] might have affected the results.

Differentiation between different bacteria becomes more reliable, if fluorescence spectra are measured at several different excitation wavelengths. The fluorescence spectra of fungal spores and b. subtilis vegetative cells e.g. are nearly identical at 266 nm excitation, but there is a distinct difference in the spectra, if excitation occurs at 351 nm [6].

Fluorescence seems to be an appropriate tool for the detection and identification of biological aerosols. Whether used for remote detection by a mobile LIDAR [7] or with a small, unattended ground sensor [8], a database of fluorescence spectra of various biological and non-biological aerosols is necessary. The aerosol fluorescence spectrometer as described below has been developed for that reason. It has been found, that the ratio of UV fluorescence to IR backscatter is characteristic for some biological aerosols [9]. The developed instrument measures backscatter coefficients for infrared light.

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2 Aerosol Generator

2.1 Bacteria

Bacteria are dispersed in a physiological aqueous sodium chloride solution, containing 1 to 10 mass percent bacteria and being stirred continuously. The bacteria had been washed and centrifuged before three times, in order to avoid rests of the nutrient growth material in the suspension. Small droplets of the suspension are generated with a PALAS AGF 10.0 aerosol generator, which uses a pneumatic atomizing nozzle and a cyclone-type classifier, holding back droplets above 5 µm size and bringing them back to the suspension. Losses of water in the nozzle and in the classifier due to evaporation are replaced by pure water, added by a peristaltic pump. The overwhelming majority of the droplets remaining in the aerosol contain no more than one bacterium, which is necessary to generate an aerosol of separate bacteria.

The aerosol of droplets flows with a velocity of 2.8 m/s through a tube of 1 m length and 15 mm diameter, being held at constant wall temperature of 40 centigrades in a heat exchanger, where the droplets evaporate completely. The aerosol is surrounded by dry sheath air of 40 centigrades and flows upwards through the measuring volume, where the optical characterization occurs. Further details are described in [10]. Typical concentrations are some 10^{10} bacteria/m^3.

2.2 Pollen

Pollen are too big for the droplet aerosol generator. They are aerosolized from dry powder in a PALAS RGB 1000, typical concentrations are some10^8 pollen/m^3.

3 Optical set-up for fluorescence measurement

Ultraviolet and visible light for the excitation of fluorescence of the aerosol is provided by a Xenon lamp (Osram XBO 450 W/1), which is focused on the entrance slit of a double monochromator (Yvon Jobin GEMINI-180, holographic gratings blazed at 250 nm). The further path of the light is shown in figure 1. The exit slit of the monochromator is imaged into the center of the aerosol flow by two off-axix parabolic reflectors P1 and P2 (Melles Griot 02PDA017/028). The spherical reflector S1 reflects passing light and increases the illumination of the aerosol. The fluorescent light from the aerosol is collected by the spherical reflector S2 and the parabolic reflectors P3 and P4 and enters the entrance slit of the fluorescence spectrometer (Yvon Jobin GEMINI-180, holographic gratings blazed at 300 nm). The intensity of the fluorescence spectrum is measured by a Peltier-cooled photomultiplier Hamamatsu R4220P in photon counting mode. More details as additional UV cut-off filters and continuous monitoring of the intensity of the excitating light are described in [10]. The complete fluorescence spectrometer runs computer-controlled (Yvon Jobin SPECTRAMAX software). Excitation wavelengths range from 220 to 570 nm, fluorescence can be detected from 250 to 650 nm.

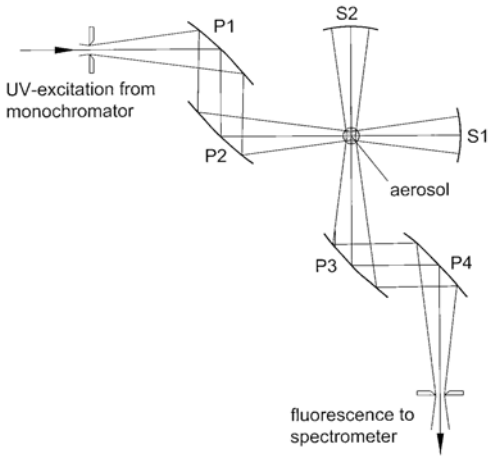

Figure 1:
Optical components for the determination of fluorescence cross-sections of biological aerosols. Aerosol flow is orthogonal to the projection plane

For the determination of the fluorescence cross-section spectra of the aerosol, the following data are necessary

1. intensity of the excitation light as a function of wavelength,
2. ratio of collected fluorescence to total emitted fluorescence,
3. sensitivity of the fluorescence spectrometer i.e. relation between intensity and count rate of the detector as a function of wavelength,
4. number of fluorescing particles,
5. measured count rates at different fluorescence wavelength.

The data for items 1. to 3. are obtained by calibration as described in [10]. Since the measuring volume of the aerosol is defined by the images of two slits, the number of the fluorescing particles can be calculated from the concentration of the aerosol, which is determined by periodic sampling. With the measurement of backscatter in the infrared which is

described below, the aerosol concentration can be determined continuously from the backscatter signal. This option, however, will be available later, since problems combining two software packages have to be solved before.

4 Optical set-up for backscatter measurement

The determination of the backscatter coefficients of the aerosol particles occurs at the same aerosol as the fluorescence measurement. As shown in figure 2, the aerosol flows upwards and is surrounded by filtered sheath air to avoid contamination of the optical components. The UV excitation light comes from left, the fluorescence goes to the front and the IR backscatter measurement occurs under an angle of 45 degrees between both.

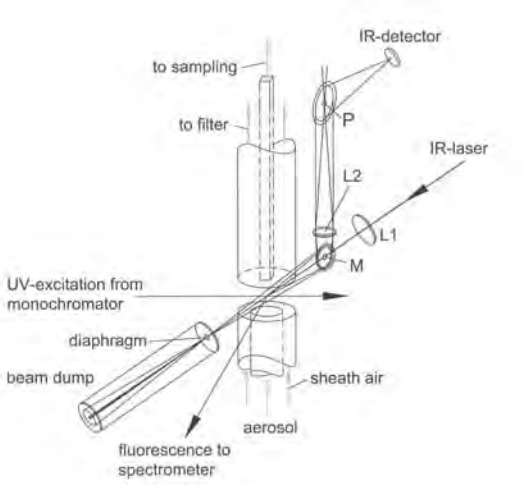

Figure 2:
Optical components for the determination of backscatter cross-sections of biological aerosols

The problem had to be solved, that the intensity of the backscattered light from the aerosol is less than a millionths of the intensity of the illuminating IR laser beam. Any scattered light of the laser beam not coming from the aerosol had to be avoided. The laser beam is focused by lens L1 on a small diaphragm in front of a beam dump. (10 mW cw-laser, 1064 nm wavelength, 1.1 mm beam diameter, 1.2 mrad divergence, LCL-LCM-T-02ccs, LASER 2000. The laser beam is modulated at a frequency of 85 Hz by a mechanical chopper.) The beam passes a 4 mm-bore in the plane mirror M before it meets the aerosol. The backscattered light from the aerosol is collected by M and directed to lens L2. This lens images the diaphragm into the center of a 8 mm-bore in a off-axis parabolic reflector P, i.e. any remaining light backscattered from the beam dump disappears through this bore. The optical system consisting of M, L2 and P images the aerosol into a IR detector (liquid nitrogen-cooled germanium diode with integrated preamplifier, Hamamatsu B7753). The electrical signal of the detector is stored in a PC via a 16 bit data acquisition board (DT3016 Data Translation, HP-VEE software). Fluorescence and IR-backscatter measurements required separate computers. To improve the accuracy of the backscatter measurement, the aerosol flow is periodically replaced by clean air flow and the difference between both signals is determined by a digital lock-in-technique.

For calibration of the instrument, a suspension of polystyrene particles of 0.95 µm diameter has been used, their backscatter coefficient being known from Mie theory.

5 Experimental results

Figures 3 to 5 show scanning electron micrographs of two different kinds of bacteria and of dactylis glomerata pollen. The particles had been sampled during the measurement of fluorescence spectra and backscatter coefficients. The size of bacteria is about one micrometer, the pollen are more than 20 times bigger.

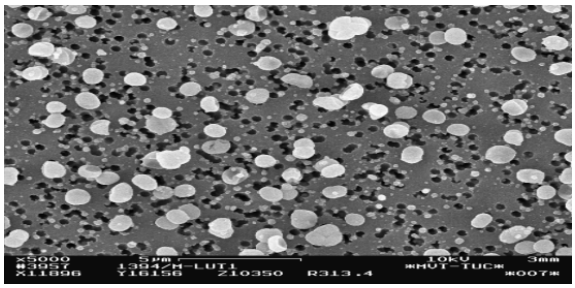

Figure 3:
SEM image of micrococcus luteus. The black objects are holes in the membrane; the small particles (< 0.3 µm) are salt crystals. Some of the micrococcus luteus bacteria can be seen to have been in the state of fission

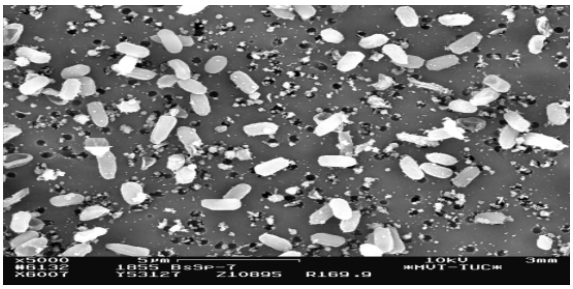

Figure 4:
SEM image of bacillus subtilis var. niger spores

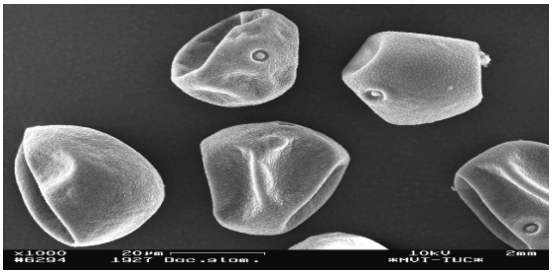

Figure 5:
SEM image of dactylis glomerata pollen

The fluorescence cross section f is defined as

$$f(\lambda_1,\lambda_2)\,\Delta\lambda_2 = P(\lambda_1,\lambda_2) / (N\, I(\lambda_1))$$

where $\Delta\lambda_2$ is the spectral width of the measured fluorescence light, P the power of fluorescence light emitted from N bacteria in the wavelength range $\lambda_2 \ldots \lambda_2 + \Delta\lambda_2$, if the bacteria are excited with light of intensity I and wavelength λ_1.

In figure 6 the measured fluorescence cross-section spectra of micrococcus luteus are plotted for four different excitation wavelengths. In figure 7 the normalized fluorescence cross section spectra of two different kinds of bacteria are compared at two different excitation wavelengths. Figure 8 shows the fluorescence cross section spectra of dactylis glomerata pollen excited at three different wavelengths and figure 9 compares relative fluorescence spectra of four different pollen.

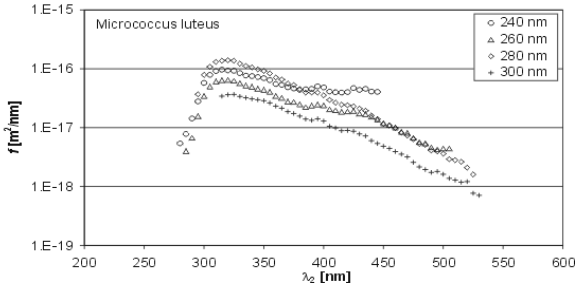

Figure 6:
Fluorescence cross-section spectra of micrococcus luteus, excitation wavelength λ_1 = 240, 260, 280 and 300 nm

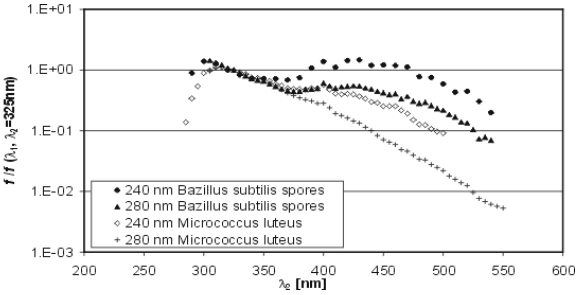

Figure 7:
Relative fluorescence spectra of micrococcus luteus and b. subtilis var. niger spores at excitation wavelength λ_1 = 240 and 280 nm

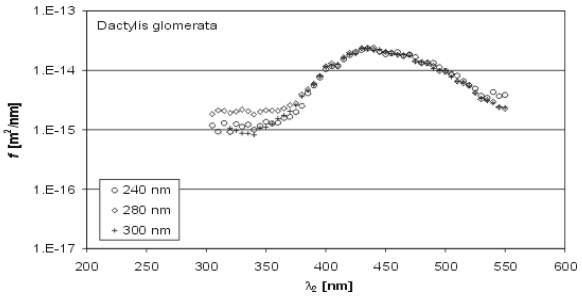

Figure 8:
Fluorescence cross-section spectra of dactylis glomerata pollen at excitation wavelength λ_1 = 240, 280 and 300 nm

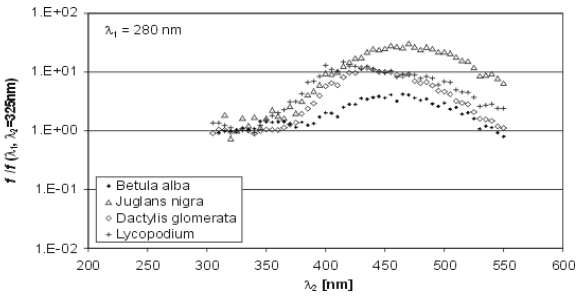

Figure 9:
Relative fluorescence spectra of four diffverent pollen resp. spores, excitation wavelength λ_1 = 280 nm

The backscatter coefficient j of a particle is defined by

$$I = N\, I_0\, \lambda^2/(2\pi r)^2\, j$$

where I_0 is the intensity and λ the wavelength of the light illuminating the aerosol consisting of N particles and I the intensity of the backscattered light at distance r from the aerosol. The wavelength of the light used for backscatter measurements was 1064 nm.

Following backscatter coefficients have been measured

Micrococcus luteus	j = 0.8
Bacillus subtilis spores	j = 0.8
Dactylis glomerata	j = 1.9 10^3

6 Discussion

The fluorescence cross-section spectra of the investigated bacteria are like fingerprints which characterize them. Bacteria have a maximum of fluorescence between 300 and 350 nm which corresponds to the maximum fluorescence of tryptophane [11]. But the influence of the excitating wavelength on fluorescence shows, that also other substances contribute to fluorescence. At shorter excitation wavelengths, the maximum becomes less pronounced, and bacillus subtilis spores show a second maximum between 400 and 500 nm if excitated at 240 nm. Pollen, however, show a weak influence of excitation wavelength on fluorescence spectra as the example dactylis glomerata showed. A pronounced maximum was found at about 425 nm, independent on excitating wavelength. This behaviour is not typical for other pollen, as figure 9 shows. More experiments on bacteria and pollen are necessary to find out, how well fluorescence cross-section spectra are suitable as indicators for specific biological aerosols. The IR-backscatter coefficients did not differ much for the different bacteria. As expected, it was orders of magnitude bigger for pollen. At their maxima the fluorescence cross-sections of dactylis glomerata and micrococcus luteus differed by a factor of about 300, their backscatter coefficients differing by a factor of 2000. This confirms the idea [9] that the ratio of fluorescence to backscatter contains additional information appropriate for differentiation between different biological aerosols.

7 Acknowledgement

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Lidar contribution to particulate matter (PM) measurements from agricultural operations

Krystyna Trzepla-Nabaglo [1] and Robert G. Flocchini [1]

Abstract

In California's San Joaquin Valley (SJV), agricultural operations are a highly complex but potentially significant source of PM_{10}. In late summer and fall a large fraction of PM_{10} is accredited to soil dust (Chow et al., 1992) becoming airborne due to agricultural activities.

We have used point sampler arrays to evaluate PM concentrations (vertical point sampler profiles) and calculate point sampler emission factors. However, because of many limitations associated with point sampler techniques, especially limited spatial resolution in vertical direction, the results may significantly underestimate the plume concentration and result in underestimates of emission factors (Holmén et al., 2001).

Relatively new application of lidar (Light Detection and Ranging) to PM measurements will be presented. Data collected with point samplers and CNL lidar (simultaneously) during variety of agricultural operations including disking and land planing in September 1999 will be reported. Recently developed new analytical methods will show lidar contribution to clarify if the shapes of plumes measured as three-point PM vertical profiles are representative of the average plumes recorded during the sampling period and help to determine the dust plume height which should be consider for integrating the modeled PM_{10} concentrations.

Results will be presented showing comparison between three-point PM vertical profiles (for selected tests) and averaged vertical profiles obtained from the lidar two dimensional (2D) scans. It will be shown how our field portable scanning lidar instrument can help overcome many of the limitations associated with traditional point sampling arrays allowing plume dynamics and PM_{10} fluxes to be described in detail.

Keywords: lidar, PM_{10} , agricultural dust, plume height

1. Introduction

Standard violation of PM_{10} in summer and fall (Dolislager and Motallebi, 1999) suggest strong influence of agricultural operations being the significant source of PM_{10} during this time of the year.

For ten years University of California researchers collected PM_{10} emissions data from variety of agricultural activities in SJV. Previous studies, however, represent limited number of sites from which only few quantified PM_{10} (Flocchini et al., 1994). Total suspended particulate matter (TSP) was measured instead. Many early studies used only single height PM_{10} samplers and TSP monitors at four heights. The insufficient for vertical profiles PM_{10} data (only one upwind and downwind) resulted in suspected underestimation of emission factors (Ashbaugh et al., 1997).

Current work includes upwind and downwind vertical profiles of wind speed and PM_{10} concentrations to quantify PM_{10} emission factors and lidar vertical profiles to verify plume heights and profile shapes.

Lidar technique as a complimentary method was employed in series of field experiments to help overcome one of the major limitations of the point samplers, determining the height of the dust plume over which to integrate the PM_{10} concentrations.

Data presented in this paper refer to a series of five valid tests conducted when point sampler data and lidar data were collected simultaneously. The results of recently developed lidar data reduction strategies are used to verify the plume heights measured with point samplers (three-point PM_{10} vertical profiles). The comparison of the lidar and point sampler results lead to some observations about validity of calculated emission factors.

2. Experimental Methods

2.1 Samplers location and field strategy

All the tests described in this paper were performed in San Joaquin Valley in September 1999. The actual measurements were conducted under real field conditions, during regular agricultural operations

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on the field. The group of five PM tests (PM99-065, 99-067, 99-068, 99-070 and 99-071) was analyzed below. The data was collected on September 8th and 9th during land planing on field 18E in Stone Land. The duration of the tests varied from 1 to 1.5 hours. Location of tractor with implement, lidar and PM towers is shown in figure 1.

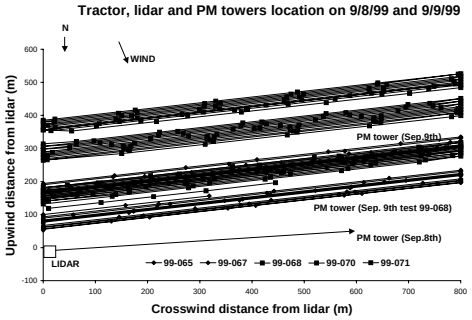

Figure 1:
Map view of tractor, lidar and PM towers locations during tests on September 8th and 9th. The tractor paths (corresponding to individual tests) were determined with laser rangefinder. The 2D vertical scans were collected upwind of PM tower as the arrow from the lidar indicates. Upwind tower (not shown) was located at coordinates (0, 1057). Stationary downwind tower (not shown) was located at coordinates (645, -195).

2.2 Tractor distance

All the tests have corresponding information about location of the tractor on the field during test period. The rangefinder (Laser Atlanta) was used independently to record the time, distance and bearing to the tractor from observer located on the West site of the field. Data was taken every 1-2 min.

2.3 Lidar (light detection and ranging)

Since its construction in 1997, the backscatter lidar instrument in Crocker Nuclear Laboratory, UC Davis has been employed in series of field studies. The greatest strength of lidar instrument is in estimating height and spread of an elevated plume providing useful information on PM plume variability over time (size, shape, etc.).

In UCD miniature elastic lidar, previously described (Holmen, 1998), light scattered back towards the lidar instrument from molecules and particles in the atmosphere is collected by a telescope and measured with a photodetector. The detector signal is digitized and analyzed by a computer to create a real-time detailed image of aerosol concentrations within the scanned region.

Specifications for the UCD miniature elastic lidar are given in Table 1 and a schematic of the system is shown in figure 2. Unlike previous lidar instruments, this lidar is capable of fast scanning, is lightweight and field portable, has low electrical power requirements, and its simple design enables effortless field operation in almost any location.

The qualitative measurements of relative PM backscatter were obtained with vertical two-dimensional (2D) scans. These measurements were analyzed to obtain vertical profiles of lidar data by averaging the lidar signal at 2 m height intervals over a specific range interval. The range interval corresponded to the location of the PM tower in the field, plus/minus 20 m on each site of the PM tower. For example, if tower was located 600 m from the lidar, the lidar vertical scans with plumes in the range of 580 m to 620m were taken under consideration for averaging.

For the same range the background vertical profiles were obtained from the scans when the tractor was not traversing the field.

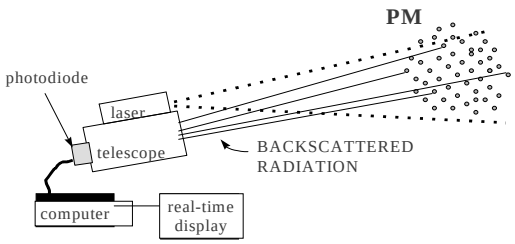

Figure 2:
Schematic of elastic backscatter lidar.

Table 1:
UCD Miniature Elastic Lidar

Specifications
• 1.064 micron, 50 Hz, 100 mJ pulsed Nd:YAG laser (light source)
• 26 cm f/10 Cassegrain telescope (light receiver)
• IR-enhanced silicon avalanche photodiode (detector mounted at telescope focal point)
• 12-bit 60 MHz digitizer
• computer-controlled scanning capabilities: 180° horizontally, 90° vertically
• field portable: overall dimensions (L x W x H) are 1m x 1m x 2m (~100 kg)
Practical Application
• detects atmospheric aerosols approx. 0.5 - 10 micron diameter
• 0.2 second time resolution
• range resolution of ~5 meters
• map of particulate matter to 6-8 kilometers distance (depending on field conditions)
• day and night operation

2.4 Point samplers

The modified aerosol samplers (Holmén et al., 2001) were used to collect PM_{10} from agricultural operations.

Locations of the tower in the field are shown in figure 1. When the stationary PM tower located at the downwind was to far from agricultural operation, the vertical profiles of PM were collected using the pneumatic tower mounted on the truck that was driven into the field. Notice the changes in locations of mobile PM tower (figure 1) as tractor operation moves North direction, further from stationary downwind PM tower.

The laboratory analyses of all collected PM samples (Holmén et al., 2001) allow the calculations of the plume heights, plume fluxes and emission factors for specific agricultural operation to be perform.

3. Results and Discussion

3.1 2D vertical scans: maximum plume height

During land planing on September 9th two-dimensional (2D) scans (figure 3A, B and C) were collected by scanning vertically along the downwind edge of the field.

The maximum heights of agricultural plumes were measured from these scans. Although the lidar cannot distinguish between PM generated by different sources, the plume generated by the tractor and implement was usually easily separated from the background PM because of the plume's distinctive movement across the field from one lidar scan to the next.

Three sequential images below (figure 3A, 3B and 3C) show the plume shape, intensity and location of the dust plume while a landplane traveled across the field. The tractor traversed from the West side to the East side of the field (A) and then turned back at about 800 m range (x-axis) and resumed its E to W direction (B and C).

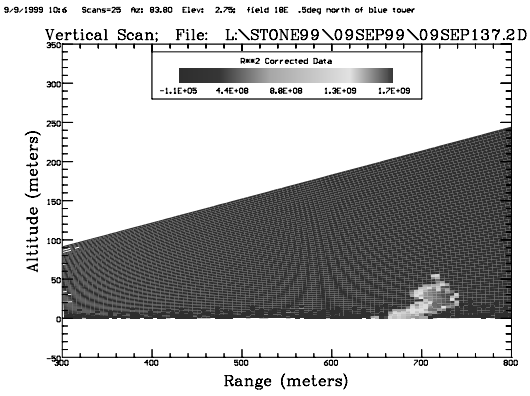

Figure 3A:
Lidar 2D vertical scan collected during land preparation. Tractor with implement (landplane) traverses across the field from West to East side. Tractor is about 700 m from the lidar. The maximum plume height (H) is about 50 m.

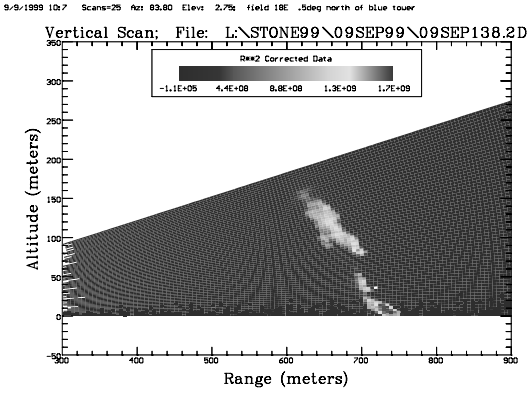

Figure 3B:
Lidar 2D vertical scan collected during land preparation. Tractor with implement traverses across the field from East to West side. Tractor is about 700 m from the lidar. The maximum plume height (H) exceeds 50 m.

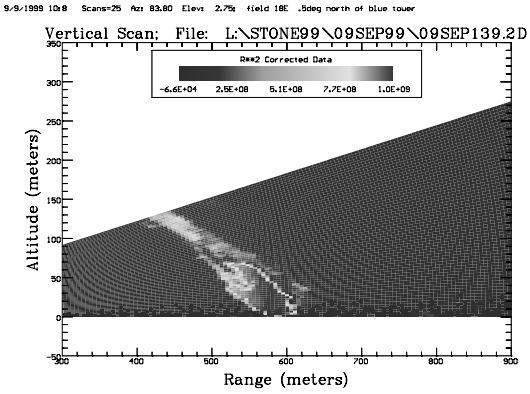

Figure 3C:
Lidar 2D vertical scan collected during land preparation. Tractor with implement traverses across the field from East to West side. Tractor is about 600 m from the lidar. Plume extended higher than the vertical limits of the lidar scan.

The tractor and implement angle measured with laser rangefinder was 80/260 degrees.

The plume drift direction (B and C) results from the change of wind direction from NW to NE.

All three images indicate that the dust plumes generated by the tractor operations reached or exceeded heights of 50 m.

3.2 PM sampler and lidar plume vertical profiles evaluation

The observations from five comprehensive tests collected with PM samplers, lidar and laser rangefinder on September 8th and 9th 1999 were used to provide better understanding of PM profiles, reasonableness of plume heights and information about factors affecting the quality of PM profile data.

All tests have upwind and downwind profiles with valid PM_{10} concentrations measurements at three heights, concurrent meteorological data and lidar vertical profiles.

During a land planing PM_{10} concentrations as a function of height were measured with filter samplers at three different heights (1, 3 and 10 m). All of the measured downwind vertical profiles showed an overall decrease in PM_{10} concentrations with increasing height. From five possible categories of downwind profile shapes (Holmén et al., 2001) analyzed group of tests represent the profile case one and case four. These case profiles seem to provide the best fit for these data. Most importantly, this seems to correlate very well with averaged lidar profiles. Because complex profile shapes were measured over limited heights with only few samplers they seem to capture actual small deviations from large overall linear decrease in concentrations with height (case four, tests 99-067, 99-068 and 99-071).

In contrast, the lidar spatial averaging over the test period and beam divergence result in relatively smooth lidar profiles compared to those measured with point samplers. This smooth averaging of lidar vertical profiles (figure 4) is the result of the high spatial resolution of the instrument in horizontal and vertical direction and the lidar scanning capabilities that allow measurements above the plume to the background levels.

The vertical profiles of averaged lidar data were calculated for both background atmosphere and plume, and selected files were averaged at the range corresponding to the location of the PM tower sampler. Background atmosphere vertical profiles were collected at the same location but when plumes due to agricultural operations were not present.

The slight decrease in the background lidar signal with time of day and between Sep. 8th and 9th was likely due to relative humidity changes and their effects on lidar response. As expected, the lidar background signal decreased with decreasing relative humidity.

The area between the lidar vertical profiles of the dust plume and the background should be proportional to the fugitive dust plume average mass during the time of the test (figure 4).

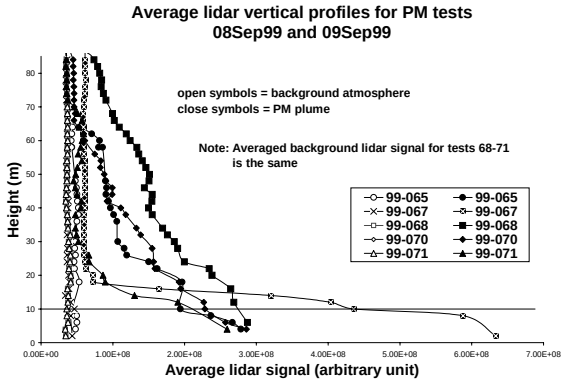

Figure 4:
Lidar vertical profiles of plumes determined by averaging lidar signal detected at the PM sampler location in height intervals of 2 meters (closed symbols). Background vertical profiles (open symbols) were collected when the plumes were not generated due to the breaks in agricultural operations.

The horizontal line at 10 m represents the highest sampler location.

As shown in figure 4 average plume heights measured with the lidar significantly exceeded the height of the point samplers (the highest point sampler was 10 m). Because of the limited number of point samplers and limitation in their sampling heights, the point samplers are often not capable of sampling the entire plume. For the averaged lidar plumes shown in figure 4 (Sep 8th and 9th) the areas representing the entire plume and the area corresponding to the height of 10 m (the highest point sampler) were compared.

It seems that for all the collected data, point samplers saw only the portion of the whole plume. Generally, the area encompassed by the point samplers varied from 21% to 65% of the entire plume captured by lidar.

Therefore, in some cases lidar correction factors of this magnitude may be necessary to apply to the point sampler results to account for the fraction of the unmeasured mass not detected by the point samplers.

It is important to note that intensity of plumes, as well as the dimensions varied during the time of the test (figure 3A, B and C) depending on changes in tractor location versus lidar sampling location and

changes in meteorological conditions (wind speed, solar radiation, relative humidity).

All lidar vertical profiles were collected at the location of mobile tower or stationary downwind tower. The location of mobile tower changed per test while stationary downwind tower stayed at the same location during the day. There were noticeable differences in PM_{10} concentrations for individual heights, even when upwind distance between point samplers was minimal. It is possible that these differences were attributed to different test-averaged distances between samplers and tractor traversing on the field, especially when the tractor was on an angle to the sampler.

It is interesting to notice registered with lidar scans high variability in plume heights over the test period (figure 5), depending on the direction of tractor travel. The changes in relative upwind distances between lidar plane and tractor seem to be one of the factors influencing plume height measurements. However, the variability in the averaged plume heights between tests performed on the same day seems to be smaller (figure 6).

Also, the lidar plume heights averaged by test in the PM sampler location and over the entire range agree fairly well. This suggests that the plume heights did not change significantly over the entire crosswind length of the tractor pass.

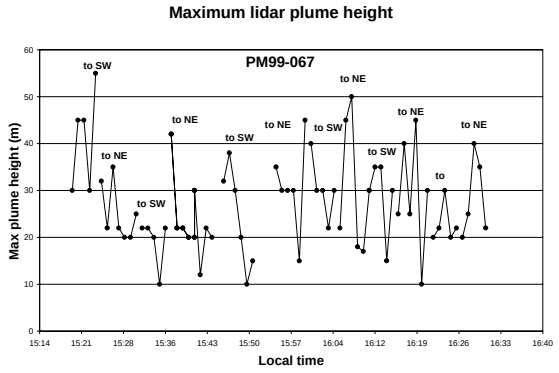

Figure 5:
Maximum plume heights recorded from the lidar 2D vertical scans during test 99-067.

Relatively low plume heights (<20 m on average) during test 99-067 may be due to increased wind speed, from 2.61 m/s during previous tests to approximately 5 m/s. Also, the effect of field-of- view could explain these significantly lower plumes during this test. Nevertheless, the data obtained from lidar vertical profiles in tower location agree fairly well with plume heights based on point samplers measurements (figure 6).

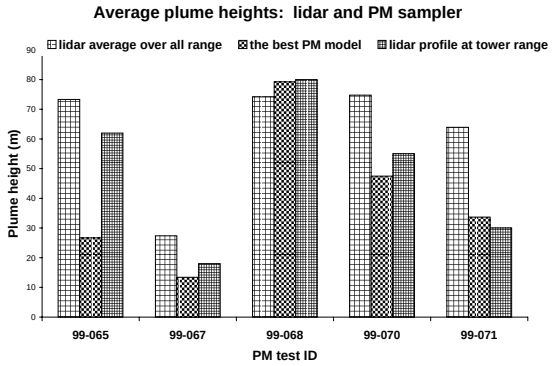

Figure 6:
Average plume heights determined by lidar (per test), by the best profile case fit to vertical PM sampler profiles and by averaging the lidar signal at the PM tower location.

It is interesting to note decreasing plume heights over the course of the day on September 9th. Again, this seems to correlate well with changes in meteorological conditions during the day, especially, increasing wind speed.

4. Summary

Results shown here from exploratory field experiments during land preparation operations in the San Joaquin Valley demonstrate the UC Davis miniature elastic lidar instrument's capabilities and the contributions lidar can make to understanding PM generation and transport from non-point sources.

Majority of plumes registered with lidar exceeded heights of 50 m. This suggests that higher PM sampler locations will be necessary to more accurately capture the whole plume. Also, additional sampler array will be valuable to produce more points for more detailed profiling resulting in more accurate plume height estimates. Because height of the plume has been a very important parameter in further calculations leading to emission factors, it is absolutely necessary to evaluate this parameter with highest accuracy.

The lidar provided important information about plume heights with much higher temporal and spatial resolution than PM samplers. The capability of the lidar instrument to scan over the plumes provided excellent independent measurements of plume heights and as the result, evaluation of vertical profiles obtained by point samplers. It is worth to note the relatively high agreement between the plume heights obtained from profiles measured with the point samplers and lidar average profiles at the point sampler location.

Collection of lidar data with future tests will continue to provide independent verification of the plume parameters, such as plume height, determined from the point sampler data.

The lidar instrument together with traditional point filter samplers can significantly improve understanding of PM emissions from agricultural operations.

5. Acknowledgements

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Measurement of particle emissions from diesel engines operating on different fuels

Hendrik Stein, Jürgen Krahl, Axel Munack, Olaf Schröder, Myriam Dutz [1]

Abstract

Diesel engines are the most common power sources in agriculture. Generally nearly as much diesel fuel as gasoline is needed in Germany for public transportation. Because diesel engines are a major source for fine particles (< 2.5 µm) and the main source for ultrafine particles (< 0.1 µm) it is necessary to determine the emissions and to characterise the particulate matter (PM). The goal of our research was to investigate and to compare particulate matter emissions from biodiesel and conventional diesel fuel. In this context biodiesel is synonymous with rape seed oil methyl ester (RME). The contribution of agriculture to PM emissions from diesel engines was about 6 % during 1996 and 2000 according to the diesel fuel consumption in Germany (Jahresbericht Mineralölwirtschaftsverband, 2000).

The Institute of Technology and Biosystems Engineering is equipped with an engine test stand to measure regulated and non-regulated emissions from engines. In detail three measurement techniques are available to detect particulate matter. The most important technique is the exhaust dilution tunnel in accordance with ECE standards with a filter unit at its end for gravimetric analysis. The second technique is a BERNER-Low-Pressure-Impactor to study the particulate mass distribution in the range from 0.015 to 16.0 µm. The third available instrument is a Scanning-Mobility-Particle-Sizer (SMPS) that detects the particle number distribution for particles with an electronical mobility diameter from 10 to 300 nm.

Three different fuels were tested in four modes of the 12-mode test (ECE-R 49). Besides RME, two common diesel fuels were chosen. In general RME showed for the mentioned measurement techniques the lowest particle emissions in the four investigated engine test points. Only partial load and rated power showed with the SMPS an increased particle number concentration between 10 and 45 nm for biodiesel versus the mineral diesel fuels. This is a remarkable result because former experiments indicated an opposite trend. Probably the progress in engine development, for example the increased injection pressure up to 1600 bar and higher, causes this positive effect. But it is necessary to point out that both the emissions for biodiesel and conventional diesel are lowered with a modern engine.

Beyond that it could be found that a low sulfur content correlates with lowered particulate matter emissions. But the effect was obviously less than the general chemical differences between mineral and renewable diesel fuel.

Keywords: Biodiesel, Diesel, Dilution Tunnel, Impactor, Particle Number Distribution, Particle Mass Distribution, SMPS, Sulfur Content

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1 Introduction

Diesel engines become more and more popular. In 2001 in Germany more than 33 % of all new sold passenger cars were diesel fueled (Jahresbericht VDA, 2001). Five years ago the percentage was only 12 % (Statistik Kraftfahrtbundesamt, 2002). The contribution of agriculture to the particle emissions from diesel engines was about 6 % during 1996 and 2000 according to the diesel fuel consumption in Germany. This underlines the importance of emission research with a special focus on particles the more because diesel engines are a major source for fine particles (< 2.5 µm) and the main source for ultrafine particles (< 0.1 µm) (Wichmann, 2002). Both kinds of particles are considered to be harmful to human health and even to be responsible for the carcinogenicity of diesel exhaust emissions (DEE). In this context it is necessary to know that ultrafine particles are obviously more toxic than fine particles regarding the same mass application rate (Wichmann, 2002; Heinrich, 1998). Furthermore, the particle number concentration is a very important property to measure and to discuss because of the high deposition capability (50 to 60 %) of ultrafine particles in the alveolar respiratory tract (Heinrich, 1998). At the Institute of Technology and Biosystems Engineering three measurement techniques are in use to detect PM emissions: A dilution tunnel to collect and measure the particle mass, a BERNER-Low-Pressure-Impactor to determine the particle mass distribution and a scanning mobibility particle sizer to measure the particle number concentration.

Besides the knowledge about particle mass and number distribution of the DEE from conventional diesel fuel it is essential to investigate which fuel parameters are probably responsible for low respectively high particle emissions. In course of a preliminary investigation three different fuels were tested in four modes of the 12-mode test (ECE-R 49). Table 1 shows the properties of the fuels. The common diesel fuels (DF 41 and DF 290) according to the European specification DIN EN 590 was delivered from Louis Dreyfus & Cie, Hannover. DF 41 is a low sulfur diesel which is sold since October 2001 at all petrol stations in Germany and fulfills the standards for Euro-IV diesel fuel. DF 290 is a high sulfur fuel according to the old standard of Euro-III diesel fuel and no longer available at the German market. Connemann Company, Leer delivered the biodiesel (RME) according to the German E DIN 51606 specification.

Table 1:
Fuel properties

Parameter	Biodiesel (RME)	Conventional Diesel Fuel (DF 41)	Conventional Diesel Fuel (DF 290)
Density at 15°C [kg/m³]	883.0	825.1	821.8
Kinematic Viscosity at 40°C [mm²/s]	4.5	2.373	2.222
Flashpoint [°C]	> 150	62.5	59.0
CFPP [°C]	- 20	- 27	- 14
Sulfur Content [mg/kg]	< 10	41	290
Carbon Residue [wt-%]	< 0.05	< 0.05	< 0.05
Cetane Number [-]	> 55	53.6	53.0
Ash Content [wt-%]	< 0.01	< 0.001	< 0.001
Water Content [mg/kg]	180	20	70
Acid Number [mg KOH/g]	0.145	0.05	
Iodine Number [-]	110		
Polycyclic Aromatic Content [vol-%]		4.9	4.2

2 Engine and Engine Test Procedure

The investigations were carried out at a modern Mercedes Benz engine type OM 904 LA with turbo charger and intercooling. The 125 kW four cylinder aggregate with a unit-pump direct injection system is often used in light duty trucks. The engine was assembled to a test bench. figure 1 shows schematically the engine test stand to detect regulated and non-regulated emissions.

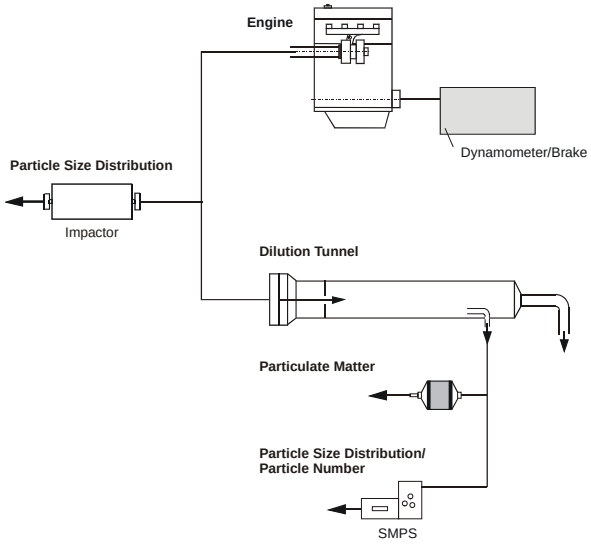

Figure 1:
Scheme of the Emission Test Stand

In figure 2 the four chosen test modes from the ECE-R 49 test are shown.

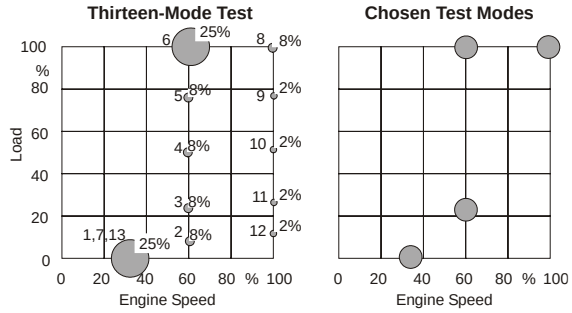

Figure 2:
13-mode test (ECE R 49) and chosen test modes

3 Measurement technique

3.1 Dilution Tunnel

The measurement of the particle mass from engine exhaust is also regulated in the ECE-R 49. It is necessary to dilute the raw exhaust and to cool it down under 52°C to assure that all volatile exhaust components are condensed on the particulate matter. After that the particle emissions were collected on PTFE-coated fiberfilm filters (T60A20, Pallflex Products). Before and after the sampling procedure the filters were weighed with a microgram balance (Sartorius M5P, ± 5 µg accuracy) to determine the emitted particulate mass. Preceding each measurement, the filters were conditioned at 22°C ± 3°C and a relative humidity (r.h.) of 45 % ± 8 % for at least 24 hours.

3.2 BERNER-Low-Pressure-Impactor

The impactor is an instrument which classifies the particulate matter into ten different size classes. Table 2 shows the characteristics of the used instrument.

Table 2:
Stages and separation diameters of the BERNER-Low-pressure-Impacter

Impactor Stages	Separation Diameter [µm]
10	8.000 – 16.00
9	4.000 – 8.000
8	2.000 – 4.000
7	1.000 – 2.000
6	0.500 – 1.000
5	0.250 – 0.500
4	0.125 – 0.250
3	0.060 – 0.125
2	0.030 – 0.060
1	0.015 – 0.030

The Berner impactor operates on the inertial behaviour of the particles. The exhaust stream is sucked through the successively arranged ten impactor stages. Starting with impactor stage number ten, the gas flows through the instrument. Every stage consists of a nozzle and an impaction plate on which a specific PM fraction is collected (figure 3). The impaction plate deflects the flow of the exhaust and causes a 90° shift in direction. Therefore a definite particle fraction with a specific aerodynamic diameter hits the impaction plate and is collected on its surface. Smaller particles will remain airborne and flow with the gas stream to the next stages.

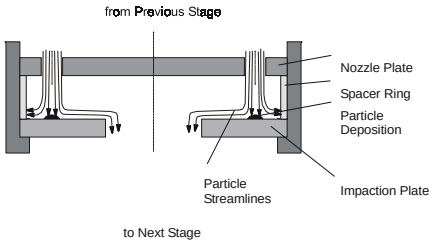

Figure 3:
Scheme of an Impactor Stage

3.3 Scanning-Mobility-Particle-Sizer (SMPS)

The principle of the SMPS is shown in figure 4.

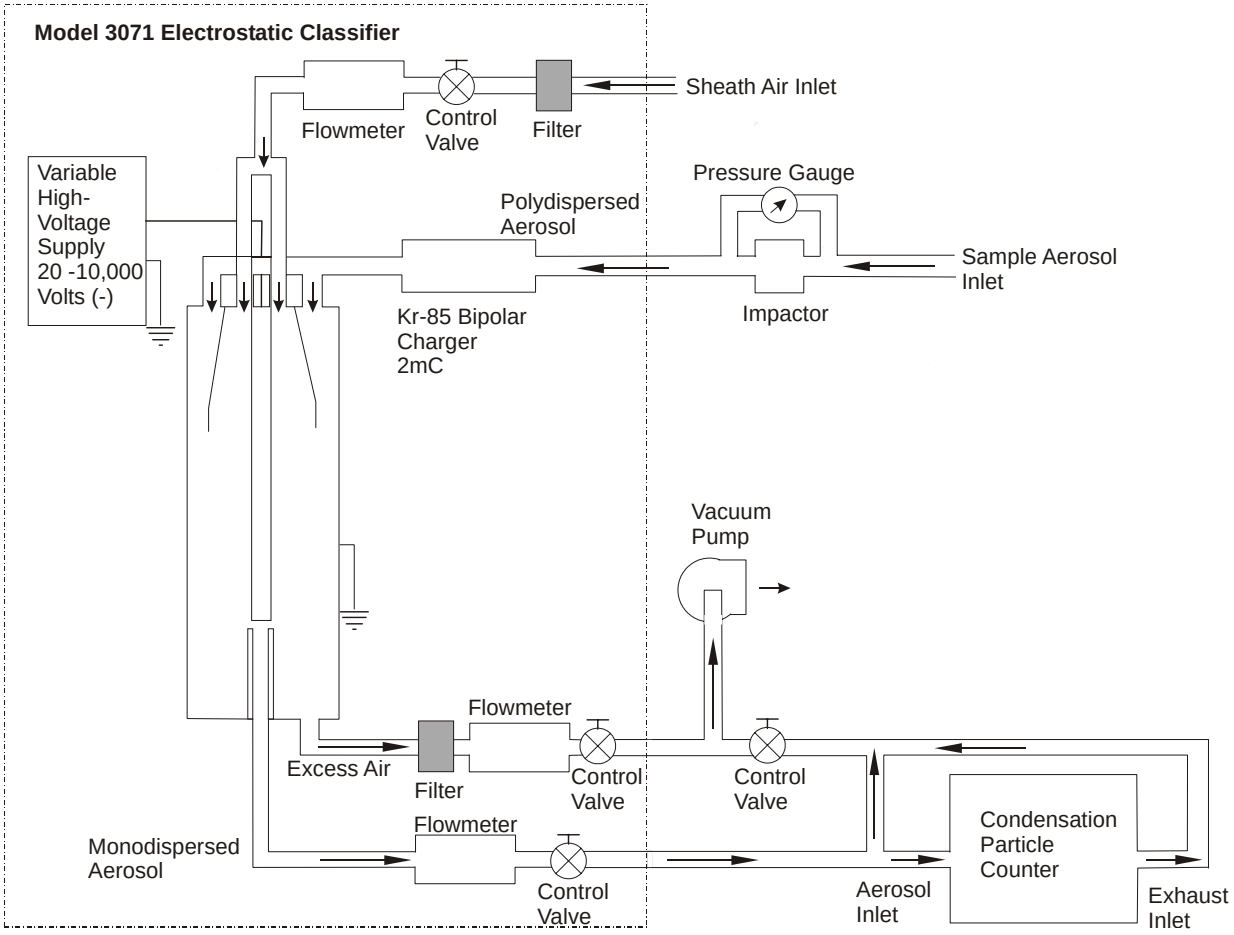

Figure 4:
Scheme of the SMPS System (Instruction Manual, TSI Incorporated)

Before the aerosol enters the instrument it passes through an inertial impactor stage to separate particles above a known diameter from the actual interesting particle fraction (10 – 300 nm). Afterwards the aerosol enters the KR-85 Bipolar Neutralizer where the exhaust stream particles collide with bipolar ions. Consequential the aerosol reaches very fast a state of equlibrium with a bipolar charge distribution. The charge distribution follows a theoretically and practically verified model developed by Wiedensohler and Fissan (1988) for particle sizes in the submicrometer regime. Then the polydispers aerosol leaves the neutralizer into the classifier zone with two concentric metal cylinders. In accordance with figure 4 the polydispers aerosol and the sheath air enter the classifier from the top and stream along the outer and inner rod. Thereby the sheath air and the sample air don't mix with each other. The inner cylinder is charged with a well-defined negative electric charge. On the other hand the outer cylinder is electrically grounded so that an electric field is created between the two rods. Thus positively charged particles from the polydispers aerosol are attracted to the inner cylinder and will be deposited there. The place where the particles depose depends on their electrical mobility and the charge of the cylinder. At the bottom of the inner rod there is a small slit where only particles with a defined electrical mobility can leave the classifier towards the condensation particle counter (CPC) as so called monodispers aerosol. During the measurement, the SMPS varies the voltage of the inner rod between 20 and 10 000 V depending on the particle sizes of interest. After the polydispers aerosol is classified into the monodispers aerosol the particle number is counted with a CPC. The aerosol flows into a heated chamber that is saturated with n-butanol. Particles and butanol vapor leave the chamber towards a condenser unit where the alcohol condenses onto the particles. The mechanism is a heterogeneous condensation. Finally the droplets' sizes are between two and three micrometer so that they can be counted optically.

4 Results and Discussion

4.1 Dilution Tunnel

Figure 5 shows exemplary the PM emissions from the tested fuels at maximum torque. It becomes obvious that RME emits less PM than the fossil fuels. The mass concentration is reduced about 40 % versus DF 290 and 35 % versus DF 41.

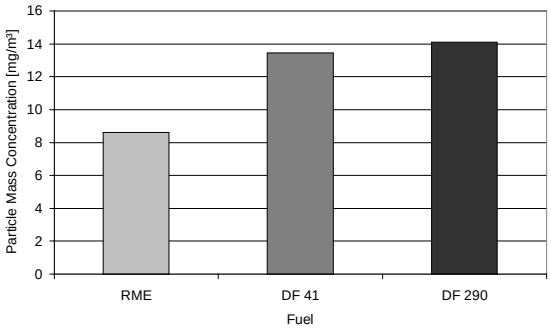

Figure 5:
Particle mass concentration for RME, DF 41 and DF 290 at maximum torque

4.2 BERNER-Low-Pressure-Impactor

Figures 6 to 9 present the impactor results for all four tested modes of the ECE R49 test cycle. The samples for the impactor were taken out of the raw exhaust gas in accordance with figure 1.

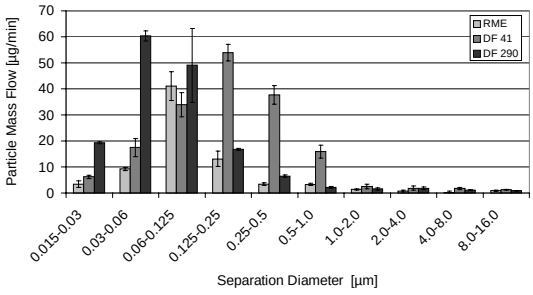

Figure 6:
Particle mass flow at idle

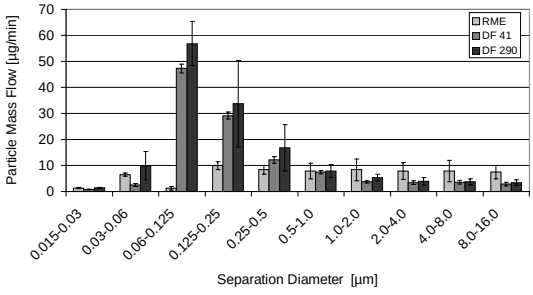

Figure 7:
Particle mass flow at partial load (25 %)

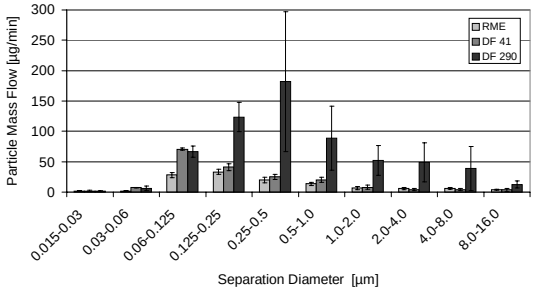

Figure 8:
Particle mass flow at maximum torque

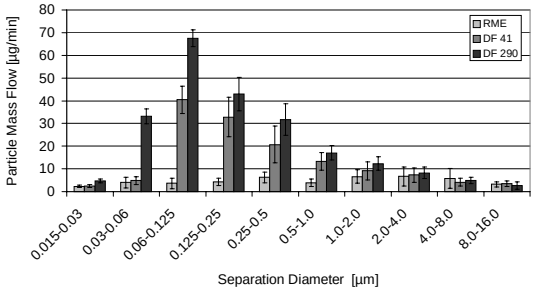

Figure 9:
Particle mass flow at rated power

For all test modes the particle mass flow is lower for RME than for the other diesel fuels. And again in comparison to figure 5, except for idle, DF 290 emits the highest particle mass per minute. This is especially very distinctive for maximum torque. Although at maximum torque the standard deviations for the DF 290 results are unproportional high, so that the picture might show too extreme differences to the other fuels, the tendency still exists.

For partial load and rated power, the maxima of the particle mass flow can be observed at separation diameters of 60 to 125 nm. Test mode idle shows an especial distribution for the three diesel fuels in comparison to the other test modes. While the maximum particle mass flow for DF 290 was found between 30 and 60 nm, DF 41 shows a maximum between 125 and 250 nm. The maximum for RME was found in between (60 – 125 nm). Although for the two diesel fuels at idle the total mass flow over all impactor stages is nearly the same, the shift to larger particle diameters for DF 41 versus DF 290 is remarkable. An opposite trend can be seen at mode maximum torque (figure 8) where the maximum for DF 41 lies between 60 and 125 nm and for DF 290 between 250 and 500 nm.

In general it is significant for the particle distributions that the preponderant part is smaller than a separation diameter of 1 µm. This fact is very important with regard to the occupational health

effects of the particles, because of their presumably increased mutagenicity.

Contrary to the expectation that the different engine loads will lead to specific shifts for the maximums, no uniform trend can be seen for the tested modes.

4.3 Scanning-Mobility-Particle-Sizer (SMPS)

Basically the particles between 10 and 300 nm can separated into two different ranges. The particles from 0 to approximately 40 nm belong to the so called nucleation mode and particles larger than 40 nm to the accumulation mode.

The following figures (figure 10 to 13) show the particle number distributions for the four test modes. They were sampled from the diluted exhaust gas (figure 1). Before the particle stream reached the SMPS it was diluted for a second time with a micro-dilution-tunnel which is not shown in figure 1.

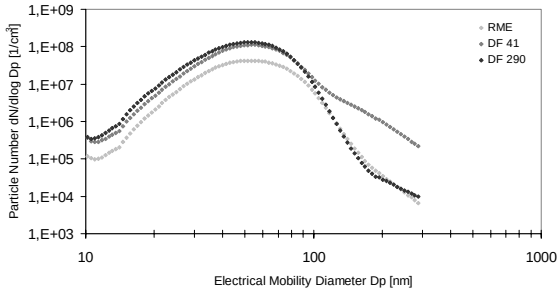

Figure 10:
Particle number distribution at idle

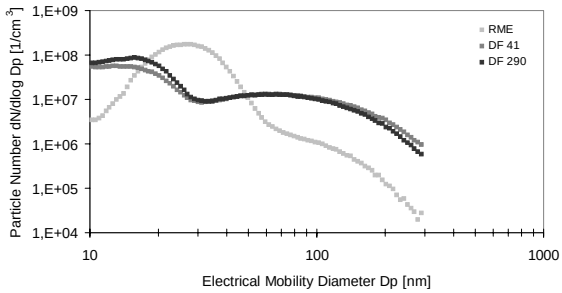

Figure 11:
Particle number distribution at partial load

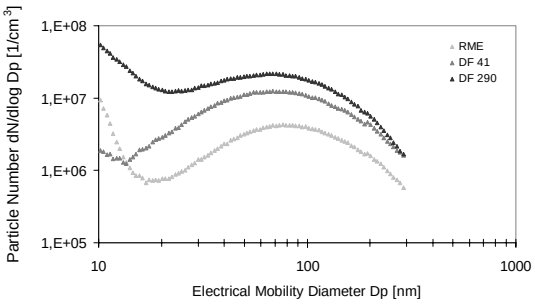

Figure 12:
Particle number distribution at maximum torque

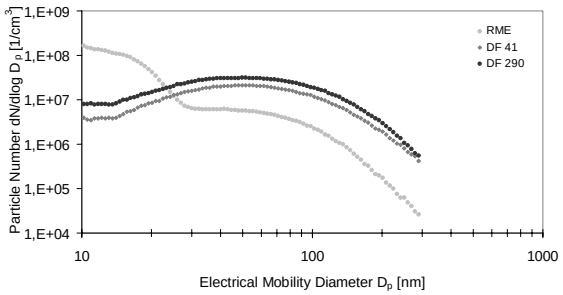

Figure 13:
Particle number distribution at rated power

DF 41 and DF 290 show a close relationship apart from maximum torque (figure 12) where DF 290 emits noticeable more particles between 10 and 30 nm.

In addition to partial load (figure 11) also maximum torque (figure 12) and rated power (figure 13) exhibit more smaller particles for RME as for the conventional diesel fuels. These particles belong to the nucleation mode particles which are very sensitive to temperature and dilution ratio changes. Experiments conducted by FEV Motorentechnik Aachen showed that it is possible to reduce these particles to nearly zero by heating the diluted sample stream (Pungs et al., 2000). Particles larger than approximately 40 nm are dedicated to the accumulation mode particles and they are insensitive to varying sampling conditions (Hall et al., 2000; Abdul-Khalek et al., 1999). In this case the temperature and dilution ratio conditions were the same for all fuels. So the differing RME nucleation mode must have an other reason.

Diesel engines are especially developed for specified diesel fuel. Biodiesel with its slightly different physical and chemical properties shows a modified combustion behaviour. In former investigations a lot of unburned RME was found to be adsorbed on the PM filters (Prieger et al., 1996). From

this it follows that the not optimised biodiesel combustion may cause additional fuel droplets which are counted by the SMPS instrument in the nucleation mode between 10 and 40 nm.

For the accumulation mode RME falls mostly below the particle number concentration of DF 41 and DF 290. The observed especial distribution at test mode idle for the impactor results cannot be repeated. Beyond that the results for the impactor and SMPS measurements are generally not comparable to each other. The measurement principles and the ways of sampling differ too much.

5 Conclusion

For all three measurement techniques the lowest particle emissions were found for RME using the four engine test modes. Only for partial load and rated power the number of nucleation mode particles was increased for biodiesel. This is remarkable because former experiments indicated an inverted trend for total particulate mass, particle mass distribution and particle number distribution (Krahl et al., 2001; Prieger et al., 1996). Probably the engine design particularly with regard to the injection technique may be responsible for that effect. While older engines injected the fuel with e.g. 380 bar (MWM 302-2) the state of the art Mercedes Benz OM 904 LA engine reaches an injection pressure of 1600 bar. The pressure difference causes a significant particulate matter reduction for the conventional diesel fuels as well as for biodiesel in particular.

Another aspect is introduced by the fuel properties (table 1), mainly the different sulfur contents of the fuels. DF 290 has the highest amount of sulfur with 290 ppm and causes nearly always the highest PM emissions. Although the sulfur content of DF 41 with 41 ppm is definitely lower, the PM emissions are only slightly lower than for DF 290. Except for maximum torque the particle number distributions are even well comparable for the two mineral diesel fuels. For RME with a sulfur content lower than 10 ppm the reduction for particulate matter (PM) is significantly higher, but the effect of sulfur content reduction is obviously less than the general chemical differences between mineral and renewable diesel fuel. In addition to the sulfur content, density, kinematic viscosity, and the flashpoint are parameters which distinguish the fossil fuels from the regenerative one.

The three different measurement techniques are good and necessary instruments for the investigation of particle emissions from diesel engines. They lead to different results with which it is possible to discuss different aspects of the emission behaviour of diesel engines operating on different fuels.

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Optical flow algorithm to quantify the two-dimensional velocity components of a visualised air jet

A. Van Brecht, K. Janssens, E. Vranken, D. Berckmans [1]

Abstract

In a ventilated space, the incoming air jet and the resulting airflow pattern play key roles in the removal or supply of heat, moisture, and removal of harmful gases and particles from or to living organisms (man, animal and plant). In this research, an optical flow algorithm was used to visualise and quantify the two-dimensional velocity components of a visualised air jet in a ventilated room. The air flow is visualised by adding smoke particles to the air. The results of the optical flow algorithm are in agreement with experimental measurements and the algorithm is a low-cost alternative technique to measure the two-dimensional velocity components. This technique might be used to measure the velocity of particles by image analysis.

1. Introduction

The main objective of a ventilation system in agricultural buildings (livestock buildings, storage rooms, greenhouses,…) is to establish satisfactory environmental conditions and air quality in the occupied zones. To realise this objective, the ventilation system must provide an efficient control of the ventilation rate and the air flow pattern (RANDALL, 1981; RANDALL and BATTAMS, 1979). Today, techniques are available to measure and control the ventilation rate continuously (BERCKMANS *et al.*, 1991; VRANKEN *et al.*, 1998).

The air flow pattern in a ventilated room is primarily determined by the momentum and trajectory of the air jet and it's mixing with the indoor air. The trajectory of a free air jet depends on the type and proximity of the inlet to the ceiling and the ratio of thermal buoyancy to the inertial forces (KOESTEL, 1955).

Analytical, numerical and experimental methods were developed to quantify and model air jet characteristics (LI *et al.*, 1993). The experimental quantification methods mainly involved velocity and temperature measurements which where also used to build advanced incomplete mixing and control volume concepts to gain further physical knowledge of imperfectly mixed processes (BERCKMANS *et al.*, 2001; BERCKMANS *et al.*, 1996; BERCKMANS *et al.*, 1993; BERCKMANS *et al.*, 1994; BERCKMANS *et al.*, 1999; BERCKMANS *et al.*, 2000; JANSSENS, 1999; JANSSENS and BERCKMANS, 1997; JANSSENS *et al.*, 2000; PRICE *et al.*, 1999; PRICE *et al.*, 1999; VRANKEN, 1993; YOUNG *et al.*, 2000; YOUNG *et al.*, 2000).

Another tool that can be used to study the characteristics of an air jet is image processing (VAN BRECHT *et al.*, 1999; VAN BRECHT *et al.*, 2000; VAN BRECHT *et al.*, 2000). An advantage of image processing over measurements with sensors can be the independency of the scale of the flow under study. By using the appropriate visualisation agent (smoke, dye,…), image processing can be used as a tool for quantitative analysis of fluid flow in many different fluids (air, water, or still other fluids) and for a very

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large range of fluid velocities. Moreover, sensors disturb the air flow while image processing, as a non-intrusive remote sensing technique, doesn't. The developed technique is inexpensive compared to other image processing techniques, and still provides good two-dimensional information of the air jet. Three dimensional quantification may be performed by using two camera's (SUN *et al.*,).

In this paper, flow visualisation with image processing is used to perform non-intrusive measurements to quantify the two-dimensional velocity components of a visualised air jet in a ventilated room.

The image processing algorithm was tested in a forced ventilated chamber, representing a section of a livestock building or a glasshouse. The magnitude of the velocity components from the optical flow algorithm is compared with velocity measurements in a two-dimensional plane in the forced ventilated chamber.

2. Materials and Methods

2.1 Laboratory test room

A laboratory test room (figure 1) (length 3 m; width 1,5 m; height 2 m; volume 9 m^3) is a mechanically ventilated chamber with a slot inlet (length 1,24 m; height 0,036 m) (1 in figure) in the left side wall 0,45 m beneath the ceiling and an asymmetrically positioned, circular air outlet (2 in figure 1) in the right side wall just above the floor. The initial direction of the supplied air with respect to the horizontal is -10°. The test room is constructed of plexiglas and is designed to create different air flow patterns by changing the ratio between thermal buoyancy and inertial forces. A second envelope chamber (length 4 m; width 2,5 m; height 3 m) in plexiglas as well is constructed around the chamber to reduce disturbing effects of varying laboratory conditions (opening doors,…). The volume of the buffering interspace is 21,875 m^3. A more detailed description of the room geometry is given by Berckmans (BERCKMANS *et al.*, 1992).

The test chamber is provided with a 3-D sensor grid consisting of 48 temperature and 24 humidity sensors. A series of five aluminium semi conductor heat sinks (3 in figure 1) and a shallow hot water reservoir (4 in figure 1) are placed at the floor to physically simulate the heat and moisture production of the biological product, plant, animal or the man. The total internal heat production is 300 Watt, the internal constant moisture production ±0,5 l_{water}/h. A mechanical ventilation system enables an accurate control of the ventilation rate in the range from 70 to 420 m^3/h or from 7,8 to 46,7 air changes an hour. In this manner, the full range of possible air jets with corrected Archimedesnumbers from 0,038 to 138 can be generated (BERCKMANS *et al.*, 1992). The resulting average inlet air speed is 0,4 to 2,6 m/s. A heat exchanger is provided to condition the supply air temperature and to simulate winter-summer and night-day outdoor conditions.

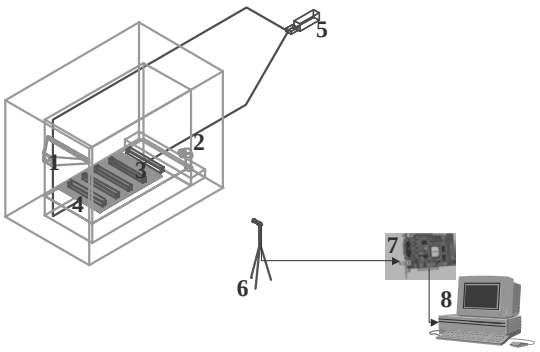

Figure 1:
Three-dimensional view of the test installation. (1) Air inlet, (2) Air outlet, (3) Aluminium semi conductor heat sinks to simulate internal heat production, (4) Shallow water reservoir with a streamer containing hot water to simulate internal moisture production, (5) Laser light sheet (at a distance of 1.50 m from the right wall), (6) CCD camera (at a distance of 4.00 m from the front wall), (7) Frame grabber, (8) Image processing computer

2.2 Flow visualisation

In general four different methods for flow visualisation can be distinguished (YANG): wall tracing, tufts, optical methods and tracer methods. The tracer method is the most popular one. Different products have been used as a tracer such as aluminium or metaldehyde particles (MURAKAMI and KATO, 1987), a smoke, a chemical, electrochemical or photochemical reaction like $TiCl_4$ (MERZKIRCH, 1987) and helium bubbles (BOON, 1978; CARPENTER *et al.*, 1972). In this study, neutrally buoyant white smoke (3-ethyleenglycol (30% of weight), propyleneglycol (30% of weight) in water) was used to visualise the air jet.

2.3 Illumination of the smoke pattern

According to Mueller (MUELLER, 1989) much attention has to be paid to the illumination of the visualised air jet. Mueller (MUELLER, 1989) describes three possible positions of the camera and the light source: when front illumination is used, the major problem is the reflection of the light on the test room; when illumination from the background is

used, it must be avoided that the camera receives direct incidence of light, but only the light diffused by the smoke pattern; illumination from the top, the left or the right side of the test chamber can always be used.

In this study, a two-dimensional illumination with a vertical laser light sheet (5 in figure 1) was used. The vertical laser light sheet is generated by a laser light source (Argon laser, 300 mWatt, mostly 514,5 nm and smaller fractions of 488 nm and 472,5 nm) in combination with a polygon scanner and had a thickness of 3 mm.

The laser sheet illumination is an accurate source of illumination since it is a two-dimensional illumination: the air jet is only visualised in the plane of the laser sheet.

For registration of the illuminated smoke pattern, a black and white CCD camera (Hitachi KP-M1E/K) was used. The camera was positioned in front of the test chamber (6 in figure 1). The back wall of the chamber was painted in black to ensure a good contrast with the white smoke pattern.

2.4 Digitalisation

A necessary step in the image analysis technique is the digitalisation of the images. With a PC-controlled digitalisation card (Matrox Frame Grabber, PIP 1024 B, 7 in figure 1) 16 consecutive images of the recorded smoke pattern are digitised. The time interval between the consecutive images is 0,7 seconds. The digitised images are 256×256 data matrices, resulting in a corresponding pixel size of 1,2×0,8 cm. The elements or pixels have a grey value varying from 0 (black) to 255 (white). Figure 2 shows the series of 16 consecutive digital images over a 11,2 seconds time period for a falling air jet.

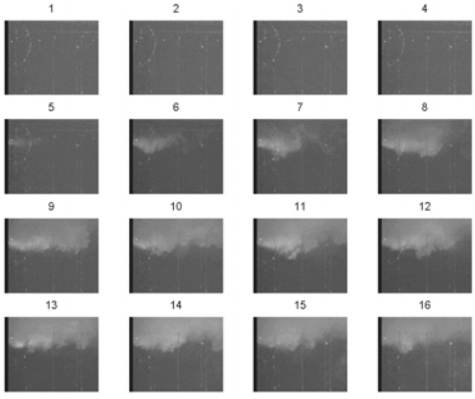

Figure 2:
Sixteen digitised images of a horizontal smoke pattern illuminated with the laser light sheet over a 10,5 seconds time period

2.5 Optical flow algorithm

The image processing method used to quantify the two-dimensional velocity components of a visualised air jet, is based on optical flow algorithm of Horn and Schunck (HORN and SCHUNCK, 1981; HORN and SCHUNCK, 1993). Optical flow is concerned with the two-dimensional motion of grey value or intensity distributions within a sequence of images. An underlying assumption is that the changing intensity patterns are in some way linked to the actual physical motion of the object. By analysing the motion of these intensity patterns, a quantitative estimate of the two-dimensional velocity distribution in the image domain may be computed.

The optical flow algorithm of Horn and Schunck (HORN and SCHUNCK, 1981) is a complex numerical algorithm that computes the difference between two consecutive images. This difference is expressed in the form of an optical flow field. Computing the optical flow vector field involves the calculation of the optical flow displacement vector $\vec{u}(x,y)$ for each image pixel (x,y):

$$\vec{u} = \begin{bmatrix} u \\ v \end{bmatrix}$$

The optical flow displacement vector $\vec{u}(x,y)$ links image pixel (x,y) with the corresponding pixel in the consecutive image. To compute the components $u = dx/dt$ and $v = dy/dt$ of the optical flow displacement vector for an image pixel (x,y), two constraints are required:

1. Consider a patch of brightness pattern that is displaced a distance δx in the x-direction and δy in the y-direction in time δt. The first constraint is that the rate of change of image brightness due to motion should be minimal. If the image brightness at the point (x,y) in the image plane at time t is denoted by $E(x,y,t)$, then the rate of change of image brightness can ε_b can be written as:

$$\varepsilon_b = E_x \cdot u + E_y \cdot v + E_t \cdot t$$

where:

$$E_x = \frac{\partial E}{\partial x}, \quad E_y = \frac{\partial E}{\partial y}, \quad E_t = \frac{\partial E}{\partial t}, \quad u = \frac{dx}{dt}, \quad v = \frac{dy}{dt}$$

2. The optical flow at a point in the image cannot be computed independently of neighbouring points without introducing a second constraint, because the vector field at each image point has two components, while the rate of change in image brightness ε_b at a point in the image plane yields only one constraint. To obtain a second constraint,

Horn and Schunck assumed that the displacement field of the brightness pattern varies smoothly almost everywhere in the image. In this case the magnitude of the gradient of the optical flow vector $\vec{u}(x, y)$ is small. One way to express this smoothness constraint is to minimise ε_c^2:

$$\varepsilon_c^2 = \left(\frac{\partial u}{\partial x}\right)^2 + \left(\frac{\partial u}{\partial y}\right)^2 + \left(\frac{\partial v}{\partial x}\right)^2 + \left(\frac{\partial v}{\partial y}\right)^2$$

Taking into account the two constraints, the computation of the optical flow vector field is reduced to the minimisation of the total error ε^2:

$$\varepsilon^2 = \iint \left(\alpha^2 \varepsilon_c^2 + \varepsilon_b^2\right) dx\, dy$$

where α is a weighing factor to account for noise in the measurements.

The minimisation of the total error ε^2 yields the following pair of equations for the optical flow vector components $u_{i,j,k}$ and $v_{i,j,k}$ that link pixel (i,j) of the kth image with the corresponding pixel in the (k+1)th image:

$$u_{i,j,k} = \overline{u_{i,j,k}} - \frac{E_x\left[E_x \overline{u_{i,j,k}} + E_y \overline{v_{i,j,k}} + E_t\right]}{(\alpha^2 + E_x^2 + E_y^2)}$$

$$v_{i,j,k} = \overline{v_{i,j,k}} - \frac{E_y\left[E_x \overline{u_{i,j,k}} + E_y \overline{v_{i,j,k}} + E_t\right]}{(\alpha^2 + E_x^2 + E_y^2)}$$

where:

$$\bar{u}_{i,j,k} = \frac{1}{6}\{u_{i-1,j,k} + u_{i,j+1,k} + u_{i+1,j,k} + u_{i,j-1,k}\} + \frac{1}{12}\{u_{i-1,j-1,k} + u_{i-1,j+1,k} + u_{i+1,j+1,k} + u_{i+1,j-1,k}\}$$

$$\bar{v}_{i,j,k} = \frac{1}{6}\{v_{i-1,j,k} + v_{i,j+1,k} + v_{i+1,j,k} + v_{i,j-1,k}\} + \frac{1}{12}\{v_{i-1,j-1,k} + v_{i-1,j+1,k} + v_{i+1,j+1,k} + v_{i+1,j-1,k}\}$$

$$E_x \approx \frac{1}{4}\{E_{i,j+1,k} - E_{i,j,k} + E_{i+1,j,k+1} - E_{i+1,j,k} + E_{i,j+1,k+1} - E_{i,j,k+1} + E_{i+1,j+1,k+1} - E_{i+1,j,k+1}\}$$

$$E_y \approx \frac{1}{4}\{E_{i+1,j,k} - E_{i,j,k} + E_{i+1,j+1,k} - E_{i,j+1,k} + E_{i+1,j,k+1} - E_{i,j,k+1} + E_{i+1,j+1,k+1} - E_{i,j+1,k+1}\}$$

$$E_t \approx \frac{1}{4}\{E_{i,j,k} - E_{i,j,k} + E_{i+1,j,k+1} - E_{i+1,j,k} + E_{i,j+1,k+1} - E_{i,j+1,k} + E_{i+1,j+1,k+1} - E_{i+1,j+1,k}\}$$

To compute the optical flow vector field between two consecutive images, the equations have to be solved for each of the pixels (i,j) of the image plane. To do so, iterative methods such as the Gauss-Seidel method can be applied (figure 3).

2.6 Experiments

Experiments were carried out in the laboratory test chamber. In the experiments, the ventilation rate was changed from 140 to 290 m³/h (15,6 – 32 air changes an hour). Each ventilation step lasted 60 min, in which the air speed was recorded in a two-dimensional grid (figure 4) of 5 horizontal and 3 vertical positions. A TSI 8455-225 hot wire sensor with an accuracy of 2%, was used to measure the air speed distribution in the ventilated test chamber. At each position, the air speed was measured 400 times with a sampling rate of 0,2 s. The air speed measurements are used to validate the optical flow field calculations

The supply air temperature was maintained at a constant level of 12°C.

The two-dimensional laser light sheet illumination was applied to illuminate the visualised air flow pattern. At each ventilation step in the experiment, the airflow pattern was visualised, filmed and digitised 10 times to have reproducible data. The optical flow algorithm processed the resulting 30 series of 16 digital images.

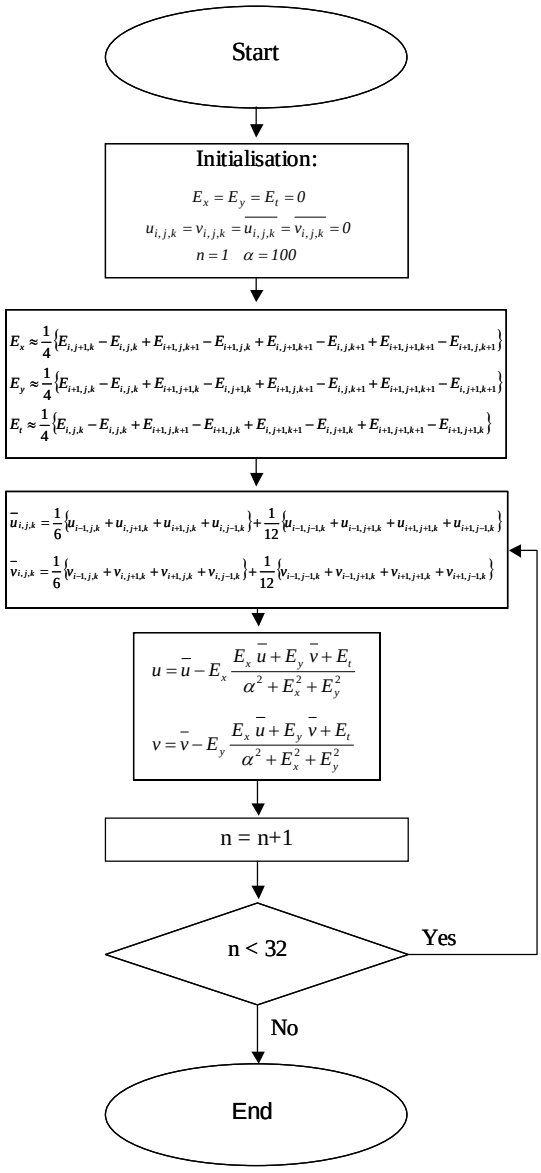

Figure 3:
Flow chart of the optical flow algorithm

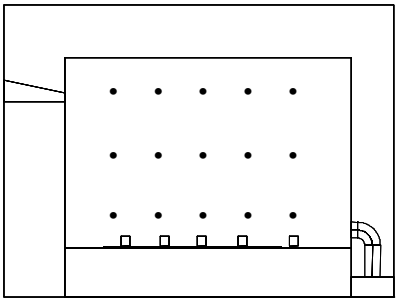

Figure 4:
Frontal view of the test installation with indication of the positions of the velocity measurements

3. Results and Discussion

The optical flow vector field calculated from the 16 images of the smoke pattern in time, the mean air speed was calculated using:

$$s_{i,j,k} = \sqrt{\left(u_{i,j,k}^{2} + v_{i,j,k}^{2}\right)}$$

In figure 5 the mean of the measured air speed with the hot wire anemometer and the mean of the air speed calculated by the optical flow algorithm are shown graphically for the ventilation rates 140, 170, 200, 230 and 290 m³/h. The maximum absolute error of the air speed calculated by the optical flow algorithm compared to the measured air speed distribution is 0,0162 m/s which is a relative error of 16%. This error may be due to changes in the sampling interval between the capture of two images. Also, the image sampling interval may be too slow.

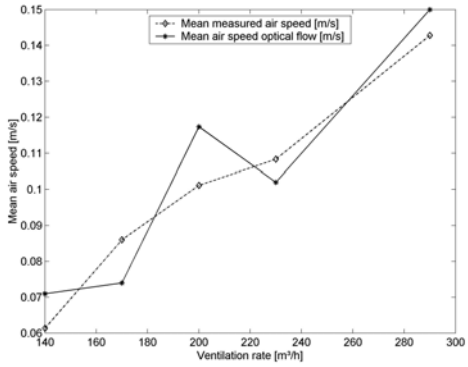

Figure 5:
Comparison of the mean of the air speed measured with a hot wire anemometer and the mean of the air speed calculated by the optical flow algorithm

Quantitatively, good agreement between the measured air speed with the hot wire anemometer and the optical flow field are shown in figure 6 for the different ventilation rates.

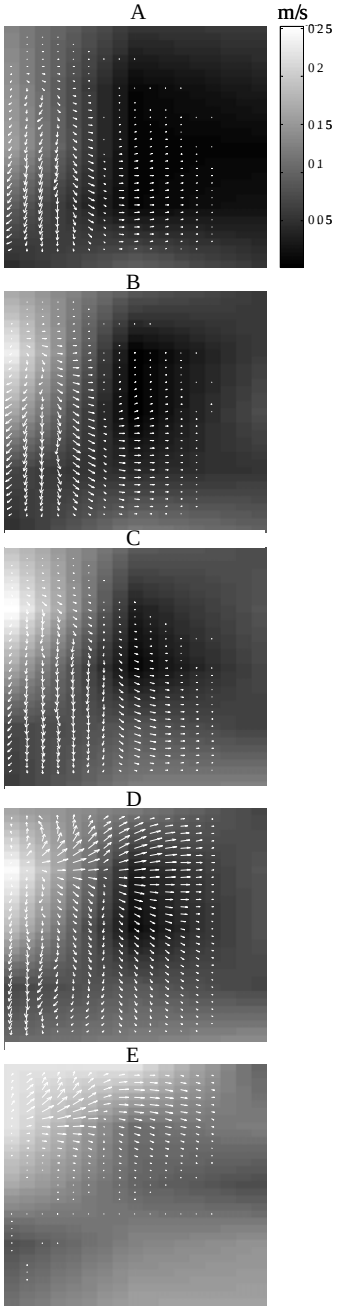

Figure 6:
Optical flow field plotted over the measured velocity magnitude distribution for an air flow rate of (A) 140 m³/h, (B) 170 m³/h, (C) 200 m³/h, (D) 230 m³/h, (E)290 m³/h.

4. Conclusions

The non-intrusive image processing method performs a low-cost quantitative analysis of air jets independent of the scale or the velocity of the flow under study. The presented algorithm may be used to perform non-intrusive air jet quantifications by the use of a laser light sheet. The maximum absolute error

of the air speed calculated by the optical flow algorithm compared to the measured air speed distribution is 0,0162 m/s which is a relative error of 16%.

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Measurement of PM_{10} and $PM_{2.5}$ emission potential from soil using the UC Davis resuspension test chamber

Omar F. Carvacho, Lowell L. Ashbaugh, Michael S. Brown, and Robert G. Flocchini [1]

Abstract

Fugitive dust emissions from natural and anthropogenic activities contribute to high concentrations of PM_{10} in California's San Joaquin Valley during the fall months. The methods currently used to estimate these emissions rely on knowledge of the dry silt content of the soil, defined as the fraction of soil that passes through a 200 mesh (75μm) screen. This parameter is not easily obtained, contributing uncertainty to the emissions estimate. Even more uncertainty is introduced in the empirical relationships between silt content and emissions.

We have constructed a dust resuspension chamber to identify geological source profiles and to investigate the potential of soil to emit dust in the PM_{10} and $PM_{2.5}$ size range. Dust is generated from soil samples in a pressurized air stream and introduced into a collection chamber. The dust is then separated into $PM_{2.5}$ using an AIHL-design $PM_{2.5}$ cyclone or into PM_{10} using a Sierra-Anderson PM_{10} inlet onto Teflon filters for gravimetric and elemental analysis. The PM_{10} or $PM_{2.5}$ dust generated from the soil can be modeled by a decaying exponential function. The model parameters are related to the inherent PM_{10} and $PM_{2.5}$ emission potential of the soil and the energy input necessary to separate the PM_{10} or $PM_{2.5}$ from the parent material.

We have optimized the chamber operating parameters to produce results that can be related to underlying soil properties. The chamber gives consistent results when used with the optimized operating parameters. We have developed a relationship between the $PM_{10}/PM_{2.5}$ index and soil textural properties using 44 soils spanning a range of soil textures. We have also tested the relationships using additional soils not used in developing them. This paper will describe the results of this research and outline a new method to estimate PM_{10} or $PM_{2.5}$ emissions from soil using readily available soil parameters

Introduction

Resuspension of soil under laboratory conditions to create dust for collection of $PM_{2.5}$ and PM_{10} permits the evaluation of $PM_{2.5}$ and PM_{10} potential of the soil under controlled conditions and the analysis of the soil independent of crop and field parameters (Carvacho, et al., 1996, 1997, 1998, 1999).

Other researchers have explored the concept of dustiness index and they concluded that the dust yield is strongly influenced by the size of the sample and the height of the drop, and that reproducibility is greater when the sample is released as a stream (BOHS, 1988; Chung and Burdett, 1994; Heitbrink, 1990a; Heitbrink, et al., 1990b; Hjemsted and Schneider, 1996; and Saxton, et al., 2000). They also found differences in dustiness depending on the sample grain size distribution.

We have found that ability of the soil to release $PM_{2.5}$ and PM_{10} under controlled conditions depends on the soil texture as defined by percent sand, clay and silt measured by the combination of wet sieving and pipette suspension which represents the soil particle size distribution for completely disaggregated soil. The soil texture parameters (the percent sand, clay and silt size fractions) are measured by combining the wet sieve analysis (<2000 μm to >50 μm) sand fraction and the pipette analysis for the clay (>2 μm) and silt (<50 μm to >2 μm) particles (Kilmer and Alexander, 1949).

The PM_{10} emissions inventory for California's San Joaquin Valley in late summer and fall includes a large fraction attributed to agricultural operations, including land preparation, harvesting, and traffic on unpaved roads (Chow, et al, 1992; Ashbaugh, et al., 1996). The emissions are currently calculated for the emissions inventory using equations that depend on silt content of the underlying soil. The silt content is defined as the fraction of soil with particles less than 75 μm physical diameter obtained by dry sieving. These particles do not necessarily have equivalent potential to emit $PM_{2.5}$ and PM_{10}. An Index of $PM_{2.5}$ and PM_{10} emissions may improve our ability to estimate $PM_{2.5}$ and PM_{10} emission from more easily measured parameters.

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Experimental

Figure 1 shows a schematic diagram of the CNL Resuspension chamber. Since we are primarily concerned with soil particles that remain suspended in ambient air, only dried soil is used to measure the maximum PM_{10} and $PM_{2.5}$ Index of the soil. Approximately 1.0 g of sieved soil material with size fraction of 0–75 µm is placed in the dust resuspension chamber, which is then sealed. An aluminum tube of 1.0 cm diameter connects the end of the dust suspension chamber to the inside of the dust collection chamber.

A measured volume of air (3.5 lpm for 15 sec) is forced through the soil sample at the base of the resuspension chamber, making a fluidizing bed. This is sufficient to suspend dust particles of approximately 50 µm aerodynamic diameter. Smaller particles are carried out of the resuspension chamber and into the collection chamber as shown in figure 1. The particles are then collected on a 47 mm Teflon filter after passing through an AIHL-design $PM_{2.5}$ cyclone or a Sierra-Anderson PM_{10} Inlet. We sample each 15 sec "puff" of dust for 15 minutes onto a single Teflon filter. We then repeat this procedure using the same sample of soil until the soil sample is depleted of PM_{10} or $PM_{2.5}$ material.

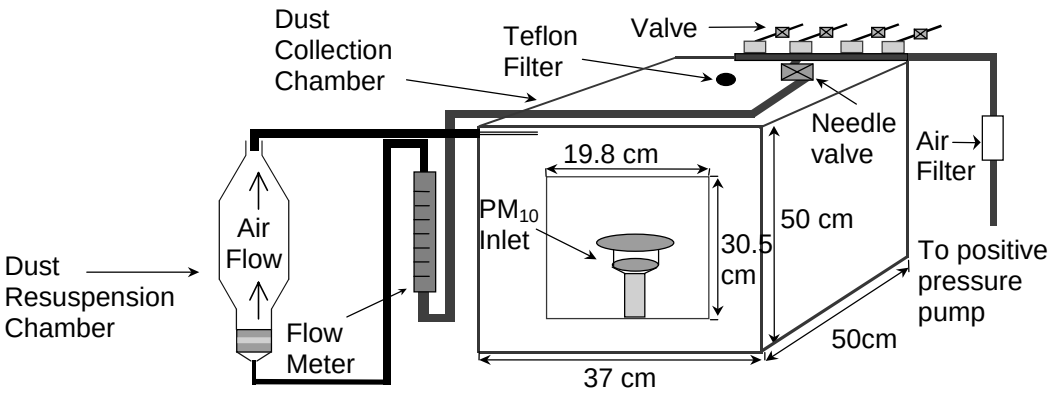

Figure 1:
Schematic of CNL resuspension and collection chamber

For this study, we collected 44 soil samples from agricultural fields, unpaved roads, paved roads, disturbed land areas, construction site, and equipment staging areas in California's San Joaquin valley. These soils spanned a wide range of texture, as shown in figure 2. Some of the agricultural soils were replicates from different parts of the same field.

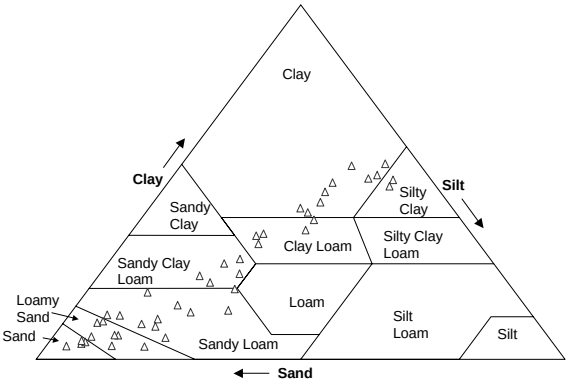

Figure 2:
Distribution of soil texture for soils analyzed

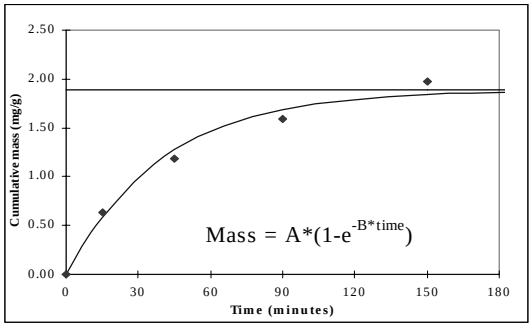

Figure 3:
PM_{10} or $PM_{2.5}$ Index curveGenerally, the unpaved roads sample was collected from agricultural roads adjacent to the fields where crop the soil sample were collected.

Both the PM_{10} and $PM_{2.5}$ index are calculated by fitting the cumulative mass CM as a function of time "t" to the equation $CM = A*(1-e^{-B*t})$ as shown in. The time parameter is the cumulative time of soil agitation (suspension) for each filter. The parameter "A" is the asymptote of the decaying exponential curve and represents the PM_{10} or $PM_{2.5}$ that would be released by repeated "puffs" if disaggregation did not occur.

Results and Discussion

Figure 2 shows the distribution of soil textures for all the soils analyzed in this study. These soil textures span the range of soil textures collected by UC Davis in the San Joaquin Valley for a study of $PM_{2.5}$ and PM_{10} emissions from agricultural operations (Ashbaugh, et al., 1996; James, et al., 1996), and also represent the range of soil textures available in the San Joaquin Valley. Sand content ranges from ~6% to over 90%, silt content ranges from less than 5% to over 40%, and clay content ranges from less than 5% to about 55%.

Figures 4-6 show the relationship between the PM_{10} and $PM_{2.5}$ Index and the standard soil texture parameters sand, silt, and clay. The $PM_{2.5}$ and the PM_{10} index is plotted for 0-75µm fraction of dry-sieved soil; both lines are shown on each figure. The $PM_{2.5}$ and PM_{10} index is the amount of $PM_{2.5}$ or PM_{10} mass, in milligrams, generated per gram of initial soil. The percent sand was measured by wet sieving, the silt, and clay were measured by pipette suspension and represent the soil particle size distribution for completely disaggregated soil. There is an excellent correlation between the $PM_{2.5}$ and PM_{10} Index and each of the three soil size fractions, but the best relationship is with clay (figure 6). The relationship with sand (figure 4)is nearly as good, and the relationship with silt has the lowest correlation . Note that the sand, silt, and clay add to 100% for each sample, so as sand increases, the amount of silt plus clay decreases. The sum of any two components (silt + clay, for example) would give the same plot as the third component (e.g. sand) but with the opposite slope. Soils with higher clay content have a higher potential to emit PM_{10} than soils with a higher sand content.

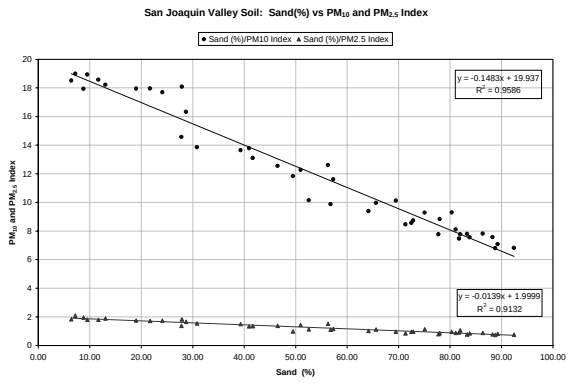

Figure 4:
San Joaquin Valley Soil: A relationship between Sand (%) by wet sieving and $PM_{2.5}$ and PM_{10} Index

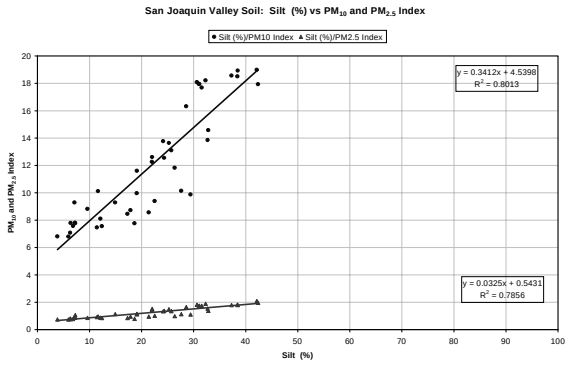

Figure 5:
San Joaquin Valley Soil: A relationship between Silt (%) by wet sieving and $PM_{2.5}$ and PM_{10} Index

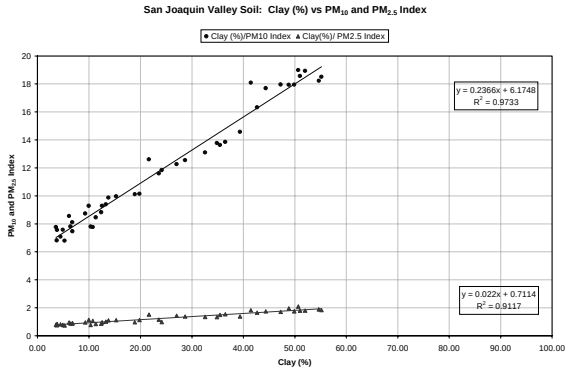

Figure 6:
San Joaquin Valley Soil: Relationship between Clay (%) by wet sieving and $PM_{2.5}$ and PM_{10} Index

Figure 7 shows the relationship between the PM_{10} Index versus the $PM_{2.5}$ Index.

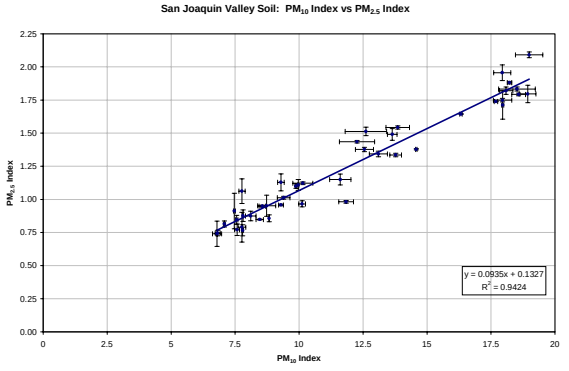

Figure 7:
Relationship between PM_{10} Index and $PM_{2.5}$ Index

The utility of any parameter used to estimate dust emissions depends on it being more easily measured than the actual emissions. At this time, the U.S. EPA

recommends using the dry silt content of soil to estimate the emissions through the use of published empirical equations. However, the dry silt content of soil is not readily available for large tracts of land and must therefore be measured for most soils. The default of 18% dry silt content suggested by EPA for unknown soils can lead to large errors in actual dust emissions. For use in agricultural emissions the soil texture is much more readily available in soil surveys published by the USDA. Furthermore, our results show that for San Joaquin Valley soils the $PM_{2.5}$ and PM_{10} Index is better correlated to the readily available soil texture, i.e. the wet sieved sand or clay content, than to the unavailable dry silt content. For these reasons, we expect the $PM_{2.5}$ and PM_{10} Index to be a better parameter to use in emission calculations.

We plan to pursue the relationship between the $PM_{2.5}$ and PM_{10} Index and measurements of dust emissions from various agricultural operations, including different soil types and with varying moisture content. If there is a good relationship between the dust emissions and the operation and soil conditions, regulatory agencies will be better able to predict dust emissions from readily available parameters.

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Size segregated particle mass concentration and chemical composition in an agrarian region in Saxony

G. Spindler [1], K. Müller, E. Brüggemann, H. Herrmann

Abstract

For a period of nine years (1993 to 2001) daily filter samples PM_{10} were collected by a high volume sampler (HV) and weekly filter samples PM_{10}, $PM_{2.5,}$ and PM_1 were collected additionally from 1999 by a low flow sampler (LF) at the IfT-research station Melpitz in the downstream plume of the Leipzig conurbation. The measuring site is located near the village Melpitz in the vicinity of the city Torgau in the river Elbe valley. The site is placed on a flat 100-years-old meadow surrounded by agricultural land.

For the whole time period the particle mass concentration PM_{10} shows a decreasing trend in the daily HV samples. Highest values have been observed in the winters before winter 1997/98. In the following winters no pronounced concentration peaks were found. A reason is the decreasing number of coal heating systems in the Leipzig conurbation. Additionally, in the last winters high pressure systems with transport of dry continental air masses and low mixing heights relatively seldom occurred.

The NO_3^-/SO_4^{2-}-ratio shows an increasing trend with seasonal variation, caused by the decreasing SO_4^{2-}-mass concentration in PM_{10} which originates in the strong decrease of SO_2 concentration, but also in NH_4NO_3-losses by evaporation from the filters during higher temperatures in summer.

From the weekly LF samples the contribution of $PM_{2.5}$ and PM_1 to PM_{10} (100%) can be estimated. Most of the $PM_{2.5}$ mass is PM_1. During summers the mass of coarse particles ($PM_{10} - PM_{2.5}$) is higher than in other seasons. One reason could be found in the occurrence of longer periods of dry ground surfaces with re-emission of crustal and biological material by turbulence, other reasons are agricultural activity and moving cars on dry roads.

The HV mass concentration measurements were integrated in a longer historical mass trend (since 1983) for Saxony.

Keywords: PM_{10}, $PM_{2..5}$, PM_1, main constituents, long term trend, seasonal variations

Zusammenfassung

Größenaufgelöste Massenkonzentration und chemische Zusammensetzung von Partikeln in einer ländlichen Region Sachsens

In neun Jahren (1993 bis 2000) wurden täglich mit einem Partikelsammler (hoher Volumenstrom, HV) PM_{10} Filterproben gesammelt. Zusätzlich erfolgte ab 1999 die Sammlung wchentlicher Proben für PM_{10}, $PM_{2,5}$ und PM_1 mit einem geringen Volumenstrom (LF) an der Forschungsstation des IfT in Melpitz im Abluftbereich des Ballungszentrums Leipzig. Der Standort befindet sich nahe dem Dorf Melpitz (Stadt Torgau) im Urstromtal der Elbe. Die Station wurde auf einer flachen 100jährigen Weide, die von Agrarland umgeben ist, errichtet. Für die gesamte Zeit zeigt die Partikelmassenkonzentration PM_{10} einen fallenden Trend in den täglichen PM_{10}-Proben (HV). Die höchsten Konzentrationen wurden in den Wintern vor dem Winter 1997/98 beobachtet. In den folgenden Wintern wurden keine auffälligen Konzentrationsmaxima registriert. Ein Grund dafür ist die sinkende Anzahl von einzelnen Braunkohlefeuerstätten im Leipziger Ballungsraum. Zusätzlich traten winterliche Hochdruckwetterlagen mit Transport trockener kontinentaler Luftmassen und niedrigen Mischungsschichthöhen seltener auf. Das NO_3^-/SO_4^{2-}Verhältnis zeigt einen fallenden Trend mit jahreszeitlichen Variationen, verursacht durch eine fallende Sulfatkonzentration, aber auch durch Verdampfung von NH_4NO_3 vom Filter bei höheren Temperaturen.

Von den wöchentlichen LF-Filtern kann der Anteil von $PM_{2,5}$ und PM_1 am PM_{10} (100%) abgeschätzt werden. Der größte Teil der $PM_{2,5}$-Masse ist PM_1. Im Sommer ist der Massenanteil des Grobstaubes (PM_{10}-$PM_{2.5}$) größer als in anderen Jahreszeiten. Eine Ursache sind im Sommer längere Trockenzeiten mit Winderosion, andere Ursachen liegen in der landwirtschaftlichen Aktivität und Aufwirbelungen verursacht von fahrenden Autos auf trockenen Straßen.

Der Verlauf der Partikelmassenkonzentration (HV) wurde in eine 1983 beginnende Messreihe für Sachsen integriert.

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Schlüsselworte: PM_{10}, $PM_{2,5}$, PM_1, Hauptbestandteile, Langzeittrend, jahreszeitliche Variabilitäten

Introduction

Up to the end of the 80-ies the region around Leipzig was one of the most polluted areas in Central Europe. Starting in 1991 the national research project SANA (Scientific Programme on Recovery of the Atmosphere above the New Federal States of Germany) was established to investigate the changes in air quality.

The collection and characterisation of environmental particulate matter is in the focus of atmospheric sciences, human health and environmental policy for several years. Nevertheless, the present knowledge on the primary formation, source identification and the effects of particulate matter is unsufficently. At the research station Melpitz (downwinds of the conurbation Leipzig-Halle-Merseburg) the input of mainly anthropogenic aerosols and other pollutants into an agrarian region was investigated from the year 1992 on beside the dry deposition of gaseous species and the wet deposition of pollutants. Beginning in summer 1992 daily samples of PM_{10} have been collected (Müller, 1999). $PM_{2.5}$ and PM_1 (Spindler et al., 1999) sampling started in 1995 and 1999, respectively. Long time series of measurements and investigations are the observational basis for the distinction of real atmospheric changes and short weather related events in the atmosphere (Heintzenberg et al., 1998).

Water soluble ions, the carbonaceous material, insoluble crustal material and water are the most important constituents. Whereas for the climatic effects the black carbon is the main component the organic material is the most interesting for human health effects and otherwise for agricultural effects the ionic constituents have important influences (Heintzenberg, 1989,. Neusüß et al., 2000).

The physical properties of particles - diameter and humidity growth - are of relevance for their lifetime, cloud processing and transportation effects (Swietlicki et al., 2000).

Characterisation of sampling site and methods

The background particle concentration was measured continuously at the IfT research station in Melpitz (Altitude 87 m, Latitude 51°32'N, Longitude 12°54'E, for detailed site description see Spindler et.al., 2001) in the downstream plume of the Leipzig conurbation. An comparison between PM_{10} HV daily samples and PM_{10} LF weekly samples looks quite well (Spindler, et al., 1999).

The HV sampler is a modified Sierra Anderson-PM_{10} sampler (Anderson Samplers Inc., USA) with quartz fibre filters (25.4 x 20.3 cm, Type MK 360, Munktell Filter, Sweden). The sampling time for each day was over 23.5 hours from 08:00 to 07:30 CET (central European time). The sampling inlet was in a height of 1.5 m over ground, the sampling volume was approximately 1340 m³.

The LF sampler is the Partisol 2000 Air Sampler (Rupprecht and Patashnik Co. Inc., USA). For the weekly filter samples PM10, PM2.5 and PM1 Teflon filters with 47 mm diameter (Millipore, Eschborn, Germany, Type 4700, 3 µm pore size) were used. The weekly sampling volume is 84 m³.

Both particle samplers realise the cut-offs with a virtual impactor. The particle mass concentration was determined for both samplers by weighing under constant conditions (50% relative humidity, temperature 20°C). After a conditioning time of 24 hours in minimum the particle mass was determined gravimetrically (Mettler AT 261 Delta Range balance, Mettler Toledo GmbH, Germany). The content of water soluble ions were determined from a quarter of the HV filters or a half of the LF filters, respectively. by standard ion chromatography procedure. Columns from Dionex, USA were used.

Long term trends in PM_{10}

Continous TSP sampling was established in 1983 by the meteorological Service of the GDR. The existing data set of Saxonian measurements was connected to the measurements of PM_{10} in Melpitz (Spindler et al., 1999). The reconstructed data (source: Sächsisches Landesamt für Umwelt und Geologie) for 1983 to 1990 present a wide scatter with mean values between 60 and 80 µg m^{-3} at rural sites. The yearly mean mass concentration decreased to 21.8 µg m^{-3} in 2000. Chemical analyses for the PM constitution do not exist for the historical data. An extrapolation of the known data on SO_2 and the data from the early 90-ies allows to speculate that sulphate was the most important ionic part of the PM. Beside the sulphate also fly ash from lignite fired power plants, industrial plants and households was a major component of the aerosol with important contents of salts, soot and metal oxides. Beginning in the 90-ies the switch off or the modernization of power plants and industrial sources as well as the individual household heating systems led to a rapid decrease of anthropogenic emissions of PM despite the increase of road traffic emissions (figure 1). Daily mean concentrations of PM_{10} reached in the winter 1993/94

15 times values above 100 µg m^{-3} whereas in the last three winters at no time. Otherwise in the last winters high pressure systems with transport of dry continental air masses and low mixing heights relatively seldom occurred.

In figure 2 the comparison of the yearly mean PM_{10} constitution from 1993 and 2000 documents the significant loss of sulphate and calcium and the relative increase of nitrate and ammonium at nearly constant concentrations. The non specified remainder seems to be constant but the carbon (organic and

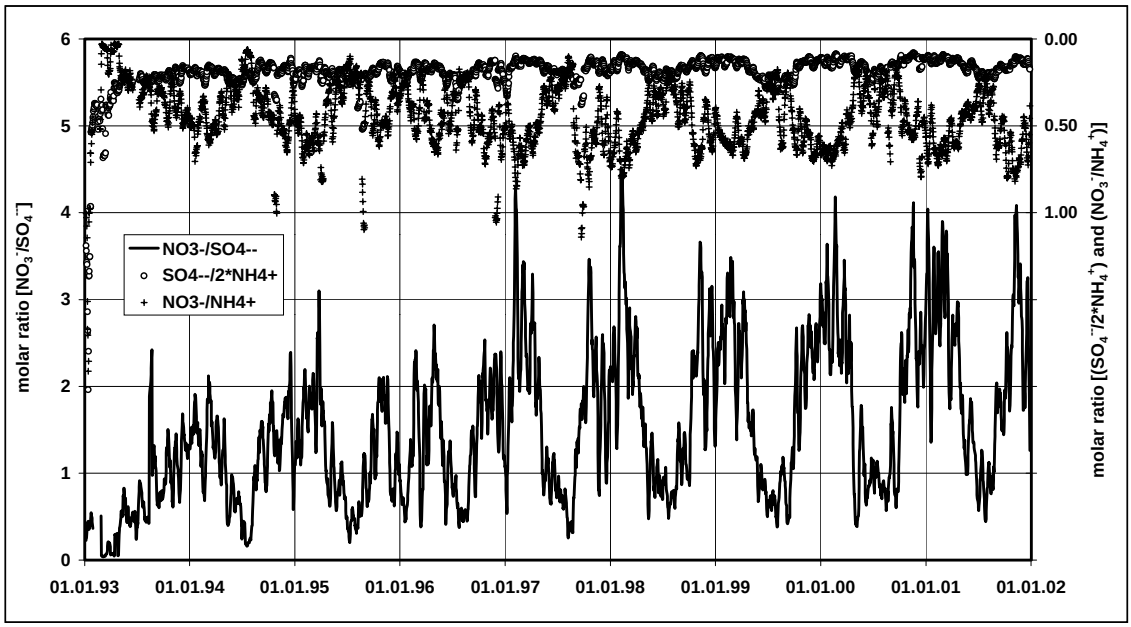

Figure 1:
PM_{10} (HV) nine year course at the Melpitz site. The ellipses highlight pronounced winter concentration peaks

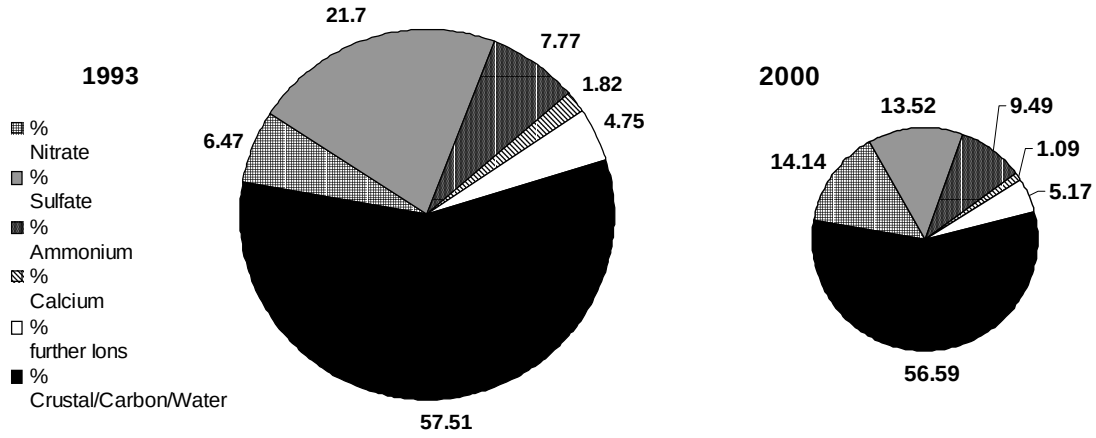

Figure 2:
Comparison of ionic components of PM_{10} in Melpitz between the years 1993 and 2000 (basis: daily samples from the HV-sampler). The area of the two circles represents the mean particle mass

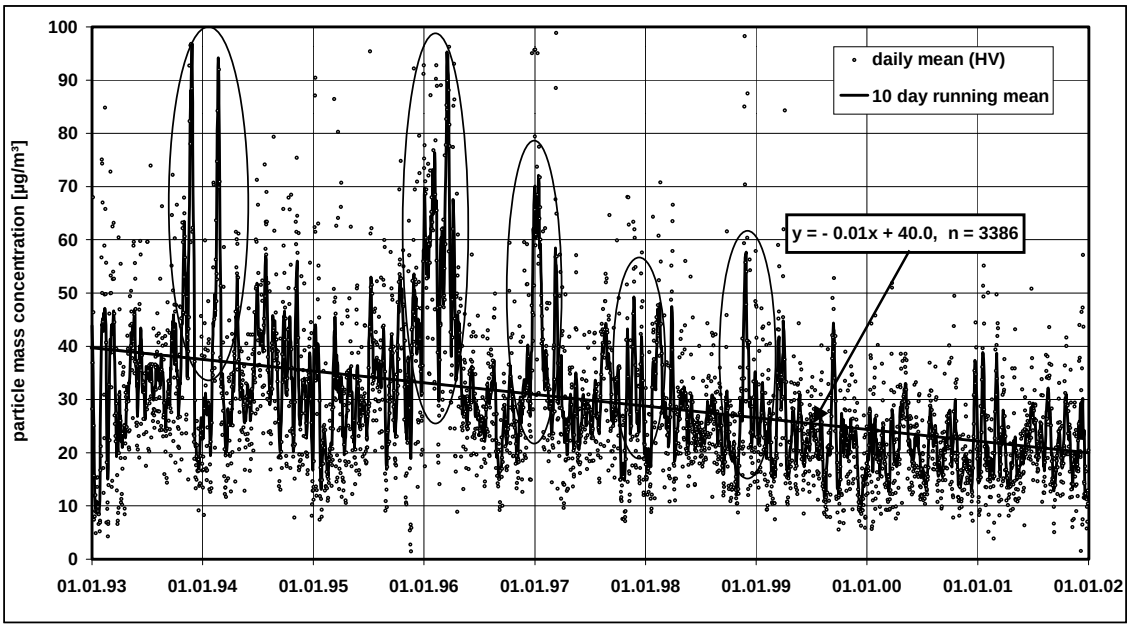

Figure 3:
Nine year course of 10 day running means of molar ratios in PM10 at the Melpitz site (the ratios for nitrate to sulphate and sulphate to ammonium plotted at the right y-axis as invers)

elemental) is decreasing while the crustal components increased by more dry and warmer summers during the last years.
From 1993 to 2000 the decrease of PM concentrations led to a decreasing deposition of mineral components as sulphur and calcium into the agricultural used land.
During the last decade a change of the most important ion from sulphate to nitrate occurred (figure 3). The seasonal variation of the nitrate content determined in the filter samples depends on the evaporation of NH_4NO_3 during higher temperatures (Schaap et al., 2001) from the used quartz fibre filters and the displacement of the gas-particle equilibrium, additionally. The ammonium concentrations are nearly stable during the seasons.

Size segregated composition PM_{10}, $PM_{2.5}$ and PM_1

The particle mass concentration and the mass of water soluble ions are available as weekly means for three particle size fractions (aerodynamic diameter PM_1: fine particles, $PM_{2.5}$-PM_1: medium particles and PM_{10}-$PM_{2.5}$: coarse particles) from the LF sampler. Table 1 lists the contribution of $PM_{2.5}$ and PM_1 to PM_{10} (100%) with a distinction between summer and winter time for 1999 to 2001. Figure 4 shows an detailed example of the mass distribution in the particle fractions for the whole year 2000 together with the daily sum of precipitation. The yearly course of the mass concentration distribution between the size fractions demonstrates the absolute maximum for the largest particles only in summer time. In summers more coarse mode particles exist, because surfaces, especially covered with short vegetation, dry faster (Klemm et al., 2002) and the absolute precipitation is lower and more intensive as in other seasons. Coarse particles caused from re-emission by turbulences and agricultural activities in the surroundings, transported only over short distances in a local area, because they have a relatively high deposition velocity. The size fraction ($PM_{2.5}$-PM_1) shows the smallest particle mass concentration over the whole year, that means the most of the $PM_{2.5}$ is PM_1.

Table 2 lists the contribution of the mean water soluble ions to the mass of the three particle fractions for all three investigated years. Sulphate and ammonium have the highest contribution to the fine particles, as result of a long range transport from natural and anthropogenic sources and particle modification by gas to particle conversion. Nitrate has the highest part of mass in the fine and medium particles. This gives a hint that combustion processes are an important source. But also the exchange of chloride during the transport from the sea to the more continental measurement site plays a roll caused by different multiphase chemical processes (ten Brink, 1998). The lowest content of water soluble ions in the coarse particles indicates re-emitted crustal material.

All discussed results of this long time study are from filter measurements, therefore in the summer

losses of ammonium nitrate (sulphate) by evaporisation caused from high temperatures and low relative humidity can reduce the mass of fine and medium particles, especially.

Table 1:
Mass contribution of $PM_{2.5}$ and PM_1 to PM_{10} (100%) distinguished between winter (October to March) and summer (April to September) for the years 1999, 2000 and 2001

	summer			winter		
PM	**1999**	**2000**	**2001**	**1999**	**2000**	**2001**
$PM_{2.5}$	56.7 %	56.8 %	65.2 %	82.0 %	76.4 %	82.8 %
PM_1	46.2 %	47.4 %	50.0 %	62.9 %	56.6 %	59.4 %

Table 2:
Relative mass contribution of the mean water soluble ions to the mass of the three particle fractions (mean for 1999 to 2001)

water soluble ion (% of particle fraction)	**particle fraction** ***(absolute mass of PM_{10} for PM_{10}=100%)***		
	PM_1 ***(54.4 %)***	**$PM_{2.5}$-PM_1** ***(15.3 %)***	**PM_{10}-$PM_{2.5}$** ***(30.3 %)***
SO_4^{2-}	21.9 %	13.9 %	5.8 %
NH_4^+	12.9 %	9.95 %	1.8 %
NO_3^-	15.9 %	21.2 %	9.1 %
sum	50.7 %	45.0 %	16.7 %

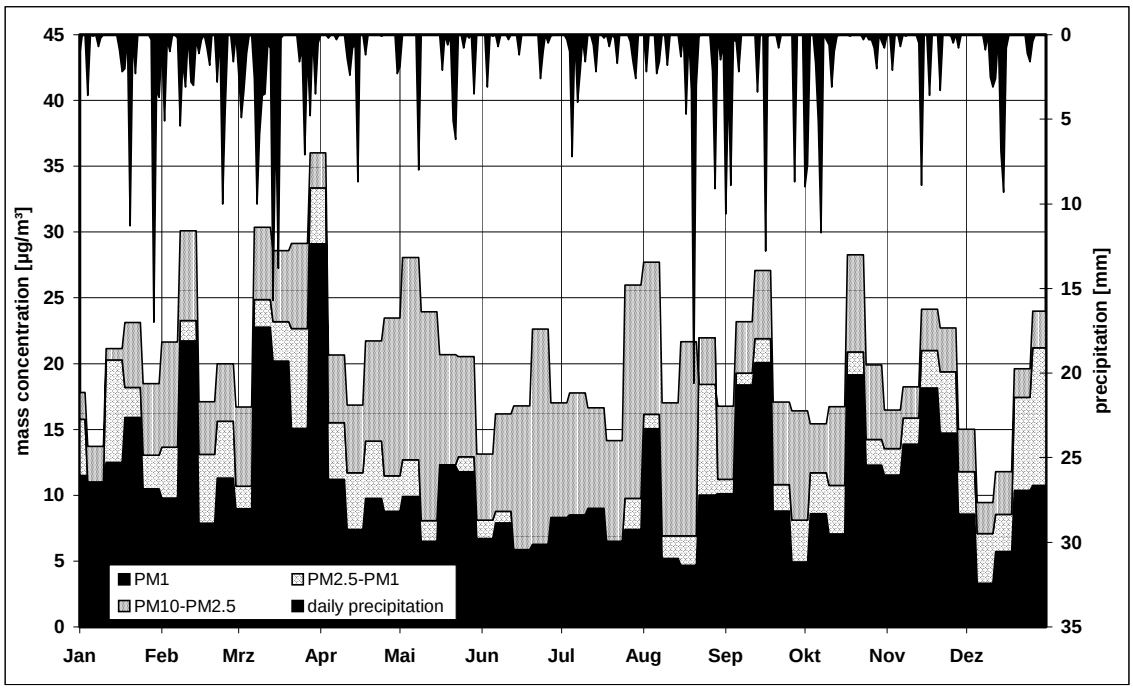

Figure 4:
Example of the mass distribution in the particle fractions for the year 2000, the daily sum of precipitation plotted at the right y-axis as invers. (week 18 interpolated and weeks 14 and 15 are calculated from daily measurements)

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Effects of bioaerosol related particulate matter on animal health

Jörg Hartung [1]

Summary

Bioaerosols are composed of viable and nonviable particles which may carry gases and which remain suspended in the air for longer periods because of their minute dimensions of between 10^{-4} and approximately 10^{2} um. Particulates in the air of animal houses originate from the feed, the litter, the manure and the animals themselves. They consist mainly of dusts and micro-organisms. Dust concentrations in piggeries range from 0.5 to 20 mg m^{-3}. Particle counts of 500,000 per litre are measured. Dust concentrations are higher in pig and poultry houses than in cattle buildings. The dust contains high amounts of proteins and carries gases, odours, microorganisms, and endotoxins; usually more than 85% of its mass consists of organic material. The level of airborne microorganisms in livestock buildings is between 100 and several thousand per litre of air. More than 80% are staphylococci and streptococci. Fungi, moulds and yeasts can form more than 1%, and coli-type bacteria about 0.5% of the total aerobic count. The behaviour of the particulates can be characterized by the processes of formation (condensation, sublimation, and dispersion) and decay (sedimentation, diffusion, and inhalation). The effect of dust and airborne microorganisms on the health of man and animals cannot be strictly separated because both form the particles that are inhaled. The aerodynamic diameter of the particles determines how deeply they can penetrate into the respiratory tract. Their impact can be described as mechanical, chemical, infectious, immunosuppressive, allergic, and toxic. Although there is evidence that high levels in animal houses may reduce production and impairs the respiratory health of farmers working regularly in animal house atmosphere, measures to reduce bioaerosol concentrations are not yet common. It seems necessary to establish scientifically based maximum levels for bioaerosols in the different livestock production systems. This would benefit both animals and farmers.

Introduction

Dust and microorganisms, polluting the air in livestock buildings, are widely considered to be principal risk factors for respiratory diseases (Clark *et al.*, 1983; Donham *et al.*, 1986; Bruce and Sommer, 1987; Wathes and Randall, 1989). Usually they occur together in an airborne state and may therefore be considered generally as particles. They can combine chemically with gases emitted into the air and also act as a carrier of odours (Hartung, 1986). However, in spite of this evidence no generally accepted maximum allowable concentrations of aerosols, particles or microorganisms in confined animal houses are defined, because the effects of airborne particles on the health and performance of livestock are still inconclusive (Perkins and Morrison, 1991). One reason for this deficiency may be the complexity of husbandry and management factors. These include: unsuitable air temperatures; air humidities and air movements; high stocking densities; low ventilation intensity; and inadequate cleaning of litter and floors (De Boer and Morrison, 1988; Perkins and Morrison, 1991). These factors can influence the formation, decay, amount, and composition of the airborne particles.

Bioaerosols, Particles and Dust

Several relevant terms are used to describe the particulates suspended in air. `Aerosols' are solid or liquid particles which remain suspended in the air for longer periods because of their minute dimensions of between 10^{-4} and approximately 10^{2} um. They can combine chemically with gases emitted into the air and these new compounds are inhaled by living organisms or can settle on them (Straubel, 1981).

`Airborne particulates` can include both solid and liquid particles. `Viable particles' are living microorganisms or any solid or liquid particles which have living microorganisms associated with them (Carpenter et al., 1986). `Non viable particles' are dispersed solid matters in gases, which arise during mechanical processes or have been stirred up. They belong to the aerosols together with smoke and fog (Henschler, 1990). `Bioaerosols` comprise odours, gases, dust, micro-organisms and other compounds

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such as endotoxins. They must be seen as a significant atmospheric contaminant and should no longer be perceived as a mere nuisance. The measurement of bioaerosol concentrations should therefore be correlated to response. This requires that particle separation, according to aerodynamic diameter, reflects the deposition pattern of the particles in the respiratory cycle. This can be described in terms of particle fractions passing size-selectors in a model filter system, which can be adopted for practical measuring procedures. Each filter of the series size-selects the particles according to aerodynamic diameter into a transmitted portion and a retained portion. The respective particle fractions can be described as nose-pharynx-larynx dust, tracheobronchial dust and alveolar dust (Henschler, 1990).

Origin and Composition of Particulates

The main aerial pollutants in animal houses are derived from similar sources. Gases are predominantly produced directly by animals and from their faeces. Microorganisms are released from animals and litter. Dust particles may originate from feed (80 to 90%), litter (55 to 68%), animal surfaces (2 to 12%), faeces (2 to 8% and, to a lesser extent, from friction against floors, walls, and other structural elements in the house (Hartung, 1986). A small amount of dust also comes from the air intake into the house (Dawson, 1990). Consequently, the particles are composed of a variety of different compounds.

From investigations of the composition of sedimentation dust in livestock housing, it is known that up to 85% of the dust consists of organic matter (Aengst, 1984). The crude protein content of pig house dust can be higher than that of the feed. This indicates that the animal makes a considerable contribution to the formation of dust. Dust from poultry housing appears to be particularly rich in protein (Hartung, 1983). Dust from horse stables was found to contain over 26% protein (Zeitler, 1988).

Dust particles also contain large numbers of microorganisms. Using suitable culture media, over 50 million colony-forming units (CFUs) of aerobically growing bacteria were detected in one gram of dust from a piggery (Aengst, 1984).

More than 80% of the airborne microorganisms found with cattle, pig, and poultry are staphylococci and streptococci. Fungi, moulds and yeasts can form more than 1%, and coli-type bacteria about 0.5% of the total aerobic count. The differentiation of fungi isolated from the air of cattle, pig, and poultry houses show regularly nine different species. The largest number of species was found in poultry houses followed by cattle and pig buildings. Some species were found to have an allergic potential.

The dust from livestock buildings contains a variety of other compounds which are potentially hazardous agents (Donham, 1989). Dust also contains potentially allergic agents, infectious microorganisms, enzymes, and toxic gases. One gram of piggery dust can absorb about 1 mg of gases such as fatty acids and phenols (Hartung, 1985). Endotoxins have been discussed widely in recent years. In pig housing, endotoxin levels of 3040 ng m^{-3}, at an average dust level of 2.1 mg m^{-3} have been found (Baekbo, 1990). Endotoxins are fragments of Gram-negative bacterial walls (lipopolysaccharides) which are known to cause allergic and immunological reactions in humans and animals.

Quantitative Findings with Regard to Dust and Microorganisms

The amount of airborne dust fluctuates greatly both in the course of a day and according to the type of animal. For example, at feeding times dust concentrations of 20 mg m^{-3} are not unusual in fattening pig houses (Nilsson, 1982). Automatic feeding, in which the animals may feed throughout the day, prevents these peaks but causes an increase in fine dust concentration due to constant movement at the feed trough (Honey and McQuitty, 1979). The number of fine dust particles (< 5µm may lie between 5000 and 3500 per litre of air (Van Wicklen and Yoder, 1988) but can reach 500,000 per litre of air in the breathing zone of farmers (Whyte *et al.*, 1993). Studies by Cermak and Ross (1978) demonstrated that the total dust concentration in poultry housing during various cleaning tasks may be up to 80 mg m^{-3}. Significant amounts of dust arise during removal of broilers and laying hens from houses and during egg collection.

In general, dust concentrations are significantly higher for pigs and poultry than in cattle houses. It is known that about 20 % (cattle), 17 % (pig) 13 % (poultry) of farmers complain about respiratory problems after work (Clark *et al.*, 1983; Pickrell, 1991, Nowak, 1998). Studies conducted at abattoirs demonstrated that up to 25% of the fattening pigs brought for slaughter in a part of northern Germany had some alteration of the lungs which stemmed from acute or prior pneumonia. This was attributed primarily to poor environmental conditions during fattening. As a cause of death in pigs, diseases of the respiratory tract are the third most important, the major causes being circulatory problems and diseases of the digestive system (Hellmers, 1986).

Airborne microorganisms, like dust, are found at higher concentrations in poultry and pig houses than in cattle buildings. Some average figures of colony-forming units per litre of air (CFUs l^{-1}) are for cattle 58 to 212, for pigs 354 to 2,000 for broiler 850 to 2,983, for layers in cages 360 to 3,781, for layers on litter 1,907 to 22,044 (up to1,000,000).

Effect of Airborne Particles

The effects of the particles in livestock buildings on human and animal health cannot simply be attributed to dust levels or the concentration of microorganisms. Effects on health are related to the complex action of particles and gases as well as the physical and psychological environment. Equally important is the immune competence of the exposed animal or man. Particulates can have effects which may be described as mechanical, infectious, immunosuppressive, allergic or toxic (Zeitler, 1988)**..** The possible health effects can be summarized as follows:

High dust levels	Mechanical irritation: overloading of lung clearance, lesions of the mucous membranes
Specific microorganisms	Infectious effect: infection by pathogens
Dust, microorganisms, and gases	Non-specific effect: defence mechanisms stressed, reduced resistance
Microorganisms and dust	Allergic effect: over-sensitivity reaction
Microorganisms and dust	Toxic effects: intoxication by bacterial/fungal toxins

The mechanical effects of high dust levels and the influence of pathogenic microorganisms are relatively easy to understand. Inhalation of large amounts of dust may cause overloading of the clearance mechanisms in the respiratory passages and mechanical irritation which facilitates the beginning of infections. High levels of dust, microorganisms or gases in the respiratory tract may lead to reduced resistance (Parry et al., 1987), particularly in animals where they may be combined with the effects of fighting within groups or unfavourable climatic conditions in the building.

Particle size is of fundamental importance to the influence of dust, irrespective of whether the inhaled particle is a grain of dust or a bacterium. The smaller the particle diameter, the deeper its point of deposition in the respiratory tract. Particles of less than 7 µm in diameter are known as alveole-accessible (Vincent and Mark, 1981; Henschler, 1990). A deciding factor in the depth of penetration is the aerodynamic diameter. At diameters of 4 to 5,um the alveolar deposition rate may be as high as 50%. It is not only the size of the dust particles which plays a part in animal health and performance. High dust concentrations seem to have a general performance-reducing effect. Carpenter et al. (1986) demonstrated that removal of a part of the airborne dust, using coarse dust filters in an air recirculation system, led to an increase in fattening pig performance. Although dust removal was only practised in the farrowing house for the first 20 days after farrowing, and the pigs were subjected to the same conditions as the control group subsequently, the animals reached their market weight up to 8 days earlier than the control group reared entirely without dust removal. Only a low level of clinically recognizable diseases occurred in both groups. However, the results could not be repeated regularly. Obviously more factors are involved than understood today.

High levels of aerial pollutants clearly influenced the health status of Danish pig herds. The results from a comparison of 44 fattening pig herds with (+AR n = 15) and without (-AR n = 29) clinically recognized atrophic rhinitis (AR) suggested a relation to the different levels of dust, bacteria, endotoxins, and ammonia which were mainly due to different ventilation rates and management practices (Baekbo, 1990). When comparing these results with a proposal for an exposure threshold given by Donham (1989), it appears that respiratory disease may be absent even under unfavourable concentrations of some of the factors (Table 1). However, the measurement technique and the sampling site in livestock buildings may have a considerable influence on the result. In horse stables, the commonly used sampling method for dust, by `area sampling' at certain sites in the animal house gives up to four times lower gravimetric readings than personal samplers which are directly attached to the animal's head (Bartz and Hartung, 1993).

A lot of attention has been given recently to endotoxins (Hartung, 1998, Hartung and Seedorf, 2000) which seem to be implicated in the pathogenesis of hypersensitive pneumonias in humans (Thelin et al., 1984). Apart from their allergic potential, they also affect the immune system (Rylander, 1986). In sensitive individuals, even very

small amounts of these lipopolysaccharides are sufficient to cause an increase in antibodies (Clark et al., 1983). In cattle and horses, allergic diseases such as rhinitis, alveolitis, and asthma are well known and are primarily associated with the use of mouldy feed, hay or straw (Siepelmeyer, 1982). The role of endotoxins in respiratory diseases of

Table 1:
Comparison of field study results with threshold values (Baekbo, 1990; Donham, 1989)

	Baeckbo		Donham
Factor	-AR	+ AR	threshold
Total dust (mg m^{-3}) (area sampling)	1.9	2.6	3.70
Endotoxin (Ug m^{-3}) (area sampling)	2.6	3.8	0.15
Ammonia (ppm)	8.0	9.0	25.0
Microorganisms (105 CFU m^{-3})	14.0	39.0	4.30

+AR and -AR, with and without clinically recognized atrophic rhinitis respectively.
25 EU = 2 ng endotoxin.

animals has not been sufficiently researched. In humans it is known that only chronic exposure over years will contribute to clinically apparent respiratory alterations like chronic bronchitis. The lifetimes of livestock may therefore be too short for damage to occur. However, even younger workers, who have spent a relatively short period in pig farming, can show temporary symptoms of irritation of the airways (Rvlander et at., 1989).

Reducing Particulate Lever

The health and production of farm animals can best be protected by improving air quality. The most important factors that influence the concentration of airborne particulates in animal houses (Hartung, 1989) are given below:

- Prevent excessive and unnecessary animal activity and unrest. For example, activities such as animal weighing should take place outside the building or should use modern methods such as image analysis which avoid moving the animals (Schofield, 1990).
- Provide low-dust distribution systems, low feed drop heights.
- Reduce dust output from dry feeds by adding oils (optional).
- Relative air humidity should not drop below 60%.
- Development of ventilation systems which minimize re-entrainment of settled dust while giving optimum ventilation rates.
- Provide a sufficient air space per animal (e.g. at least 3 m^3 per fattening pig).
- Use technical equipment such as vacuum cleaners (Nilsson, 1982) or sprinkling of water.

Bioaerosols in the air of livestock buildings can present a significant burden to the respiratory tract of humans and animals and must be considered in the context of some typical respiratory disease patterns. The effects of dust, microorganisms, gases, and toxins cannot be separated, but dust is of special significance as the carrier of these substances. Measures to reduce dust formation in the house should be given more sustained application in practice. This would be in the interests of workers, of animal welfare, and of the wider environment. There is an increasing concern about the emission of particulates from farm buildings which may cause harm to the population living in the vicinity of large animal units. There is some evidence to suggest that bacterial and fungal levels in surrounding of large livestock enterprises are higher at times than in areas where there is little or no livestock (Hartung, 1992). The amount, quality and travel distance of the compounds has yet to be investigated. The role of bioaerosols in livestock production should be given more attention in future.

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Characterization of organic dust exposure by using a human whole blood assay

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Abstract

Exposure to organic dusts or bioaerosols is associated with a variety of acute and chronic respiratory symptoms. In seeking to control organic dust-induced diseases it is necessary to identify and quantify the toxic agents in the dust.

Here we describe an in vitro assay – a human whole blood assay- that can be used to estimate the inflammatory potency of organic dust samples. With the whole blood assay the inflammation reaction that bioaerosols cause in the respiratory tract is exactly characterized. That assay is able to detect a broad spectrum of macrophage activating substances and not only endotoxins as the limulus assay.

1 Introduction

Organic dusts or bioaerosols consist of particles of biological origin and may be very complex in nature. Exposure to organic dust can induce a variety of pulmonary diseases and subjective symptoms. Some of them have an inflammatory origin, like toxic pneumonitis or chronic bronchitis. Typical occupational environments where such diseases can be observed are animal houses, waste handling facilities or cotton card rooms (e.g. Rylander, 1994). About 12 million employers are exposed to workplace bioaerosols in Germany today. Further, organic particles emitted from these facilities are supposed to cause health implications in nearby residents (e.g. Hartung, 1998).

The inflammatory reaction in the respiratory system is caused by various agents of the dust like endotoxins, glucans or tannic acid. These agents activate different cells in the lung tissue, chiefly macrophages to excrete inflammatory mediators like IL-1ß and TNF-α. The result of this activation process is the release of further cytokines, chemokines and also tissue-damaging cytotoxic mediators, which initiate and modulate the inflammation process (Hold, 1990; Rylander, 1994).

Currently risk assessment to organic dust exposure is mainly based on the determination of airborne endotoxin concentration. Airborne dust is collected by filtration, the collected dust is extracted in pyrogen-free water and the endotoxic activity in the extract is determined by using the Limulus assay. However, the value of the Limulus reaction to characterize the inflammation process in mammals is under discussion for various reasons: 1) the Limulus assay is based on the coagulation reaction of amebocytes of a crab and may not reflect the inflammation reaction in mammals, 2) the Limulus assay is limited to the detection of endotoxins and 3) the assay becomes very prone to interference by chemical or physico-chemical interaction, a serious problem in complex organic dust samples (e.g. Danuser and Monn, 1999).
In this study we compared the activity of typical components of organic dust from animal houses to coagulate Limulus Amebocytes and to induce IL-1ß in human blood macrophages to gain information about the problems described above.

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2 Material and Methods

Strains of bacterial species, which were frequently isolated from the airborne state of animal houses as well as commercial available Lipopolysaccharids (LPS) and 1-3-ß-D- glucans, were tested in the present study.

The Limulus Amebocyte Lysate- (LAL) assay QLC-1000 (Bio Whittaker, Walkersville, USA) was used to characterize the ability of these substances to agglutinate amebocytes of the horseshoe crab *Limulus polyphemus*. The ability of these substances to activate human blood macrophages was determined by using a human whole blood assay as described by Hartung and Wendel (1996). For a Control Standard Endotoxin (CSE; *E. coli* 0113:H10; Chromogenix, Italy) the Limulus assay had a detection limit of 0.05 EU/ml and the whole blood assay a limit of 0.5 EU/ml.

2. 1 Testing of bacterial strains

The origin and the culture conditions of the tested bacterial strains are listed in table 1. After cultivation the bacteria were harvested by using pyrogen-free water. These bacterial suspensions were adjusted to an optical density of 1.0 at 520 nm. From these suspensions serial dilutions were prepared (dilution factor 10). After that a heat inactivation (30 min at 80°C in a water-bath) was done. In both test systems that dilution was determined that caused a just positive reaction.

2.2 Testing of Lipopolysaccharides and 1-3-ß-D-glucans

The substances used in this study are listed in table 2. These substances were dissolved in pyrogen-free water and concentrations from 1mg/ml to 1pg/ml were investigated in both test systems. In both test systems that concentration was determined that caused a just positive reaction.

Table 1:
Origin and culture conditions of the tested bacterial strains

Strain	Origin	Culture conditions
E. coli	air, hen house	Standard I agar, aerobic, 24h, 37°C
E. coli 1	air, pig house	Standard I agar, aerobic, 24h, 37°C
Ent. agglomerans	air, hen house	Standard I Agar, aerobic, 24h, 37°C
Ent. agglomerans 1	air, hen house	Standard I Agar, aerobic, 24h, 37°C
Pseudomonas putida	air, hen house	Standard I agar, aerobic, 24h, 30°C
Pseudomonas fluorescens	air, hen house	Standard I agar, aerobic, 24h, 30°C
Acinetobacter lwoffii	air, hen house	Standard I agar, aerobic, 24h, 37°C
Bacteroides vulgatus	feces, calf	TAS agar, anaerobic, 72h, 37°C
Bacteroides fragilis	feces, calf	TAS agar, anaerobic, 72h, 37°C
Streptococcus faecalis	strain collection	Standard I agar, aerobic, 24h, 37°C
Staphylococcus xylosus	strain collection	Standard I agar, aerobic, 24h, 37°C
Clostridium perfringens	strain collection	Standard I agar, anaerobic, 24h, 37°C
Micrococcus luteus	strain collection	Standard I agar, aerobic, 24h, 37°C

3 Results

3.1 Reactivity of bacteria

The reactivity of the different bacterial strains in both test systems is given in figure 1. In the LAL assay 3 main bacterial groups can be distinguished. All Gram-positive bacteria exhibited a very low reactivity. Obligate anaerobic Gram-negative bacteria showed a medium reactivity. Aerobic and facultative anaerobic Gram-negative bacteria showed a high and very uniform reactivity in the LAL assay.

However, in the human whole blood assay the reactivity of these bacterial groups was much more heterogeneous. Within the aerobic and facultative anaerobic Gram-negative bacteria the *Enterobacteriaceae* exhibited the highest reactivity in the whole blood assay. *Neisseriaceae* and *Pseudomonadaceae* were less potent in the activation of monocytes compared to the *Enterobacteriaceae.* The reactivity of the Gram-positive bacteria was in general lower than the reactivity of the Gram-negative bacteria. However these bacteria caused an activation of monocytes in dilutions where they did not caused a coagulation of Limulus amebocytes.

3.2 Reactivity of LPS and glucans

The reactivity of different glucans and a CSE in the whole blood assay and the LAL assay is given in table 2. It becomes clear that the tested glucans are very weak activators of monocytes compared to the CSE.

However, the interpretation of the found low reactivity of the tested glucans is difficult. We could not exclude that the observed weak activity is caused by a contamination of the glucans by LPS, since the glucans reacted also positive in the Limulus assay. But it is also possible that the positive reaction in the Limulus assay is caused by an activation of the glucan-reactive factor G (Hurley, 1995).

Table 2:
Reactivity of (1-3)-ß-D-glucans and a Control Standard Endotoxin in the Limulus and the whole blood assay

Substance	Limulus assay	IL-1ß (whole blood assay)
CSE*	5 pg/ml	50 pg/ml
Curdlan	50 µg/ml	500 µg/ml
Laminarin	100 µg/ml	>1 mg/ml
Zymosan	1 µg/ml	100 µg/ml

Substances were dissolved in pyrogen free water. In the table that concentrations are shown, that caused a just positive reaction in the limulus or whole blood assay. (* CSE = Control Standard Endotoxin, *E. coli* 0113:H10, Chromogenix, Italy)

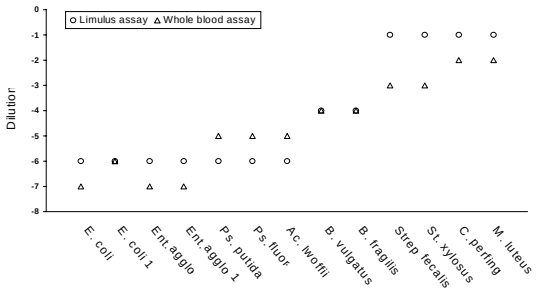

Figure 1:
Reactivity of heat-inactivated bacteria in a Limulus assay and a whole blood assay

Bacterial strains were cultured under defined conditions and harvested in pyrogen-free water. These bacterial suspensions were adjusted to an optical density of 1.0 at 520 nm and serial dilutions were prepared (Dilution factor 10). After a heat inactivation in both test systems that dilution was determined that caused a just positive reaction. (*Ent. = Enterobacter, agglo. = agglomerans, Ps. = Pseudomonas, fluor. = fluorescens, Ac. = Acinetobacter, B. = Bacteroides, Strep. = Streptococcus, St. = Staphylococcus, C. = Clostridium, M. = Micrococcus*)

Discussion

The results showed that endotoxins are the most potent activators of human monocytes as well as the most potent inducers of Limulus Amebocyte coagulation. However, the results also indicate that the reaction of humans to endotoxins of different bacterial species is much more heterogeneous as indicated by the Limulus assay. For instance the Limulus assay overrates the inflammatory activity of *Pseudomonadaceae* compared to *Enterobacteriaceae*.

Further, the results indicate that besides endotoxins also other microbial components, like components of Gram-positive bacteria, are able to activate inflammatory cells of the respiratory tract. That is in keeping with results from Larsson et al. (1999). These authors have shown that Gram-positive bacteria induce IL-6 and IL-8 production in human alveolar macrophages and epithelial cells. However, such components will not be detected by the Limulus assay. Therefore, the potential role of these components in the inflammation process induced by bioaerosols cannot be addressed by using the Limulus assay.

The whole blood assay can be easily adopted to standard sampling procedures for bioaerosols, like filtration or impingement. Exposed filters can be placed directly into the blood, special extraction procedures as for the Limulus assay are not necessary Fennrich et al., (2000).

An exposure assessment to bioaerosol based on the whole blood assay offers various advantages over the Limulus assay. Whole blood assays are able to detect a broad spectrum of macrophage activating substances and not only endotoxins. Therefore an overall exposure-monitoring should be possible, since organic dust exposures are generally mixed - different microbial components are almost always present. With the whole blood assay the inflammation reaction that bioaerosols cause in the respiratory tract is exactly characterized. Therefore we expect that exposure assessment by using the whole blood assay could also improve the quality of general risk assessment. Beside the function of an exposure marker the whole blood assay may act as a biological effect marker to determine personal susceptibility of the blood donor to special organic dusts. It should be investigated whether the proinflammatory immune response measured ex vivo by means of the whole blood assay presents a valuable predictive individual indicator in the preliminary stage of acute and chronic respiratory disorders.

Furthermore, it is possible to establish a whole blood assay not only for humans but also for other mammals of interest like horses or pigs. Exposure to

bioaerosols is also a serious problem in veterinary medicine.

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Production and disease effects of dust and ammonia on the weaner pig

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Abstract

Pigs were exposed for six weeks to controlled concentrations of airborne dust and ammonia in a single, multifactorial experiment. Production and health responses were measured to establish the tolerable concentrations of these pollutants. The treatments were dust concentration of either ≈0, 2.5, 5 or 10 mg m^{-3}. (inhalable fraction) and ammonia concentration of either ≈0, 10, 20 or 40 ppm, which are representative of commercial conditions. The experiment was carried out over 2.5 years and pigs were used in eight batches each comprising five lots of 24 pigs. Each treatment combination was replicated once and an additional control group (≈0 mg m^{-3} dust and ≈0 ppm ammonia) was included in each batch to provide a baseline. For the other four lots in each batch, the dust concentration was common while all four ammonia concentrations were used: thus the design was more sensitive to the effects of ammonia than dust.

The pigs were kept in a purpose-built facility, comprising 5 identical rooms. The pollutants were injected continuously into air supply of each room. An artificial dust was manufactured from feed, barley straw and faeces, mixed by weight in the proportions 0.5:0.1:0.4 and re-suspended in the inlet air. Ammonia was supplied from a bottle bank.

Liveweight gain and food intake were measured after three and six weeks of exposure. Observations of health were made daily and focussed on clinical signs of respiratory disease. At the end of exposure, eight pigs from each lot were killed for pathological, serological and microbiological examinations, again with am emphasis on respiratory disease.

The experiment finished in December 2001 and the complete data have just become available for statistical analysis. This is currently underway and the main findings will be presented at the conference. This paper describes the facility, the environment data and summarises the main production results.

Keywords: Dust, ammonia, weaners

1 Introduction

Housed pigs are regularly exposed to high concentrations of aerial pollutants, which result from deficiencies in building design and operation. These pollutants arise from manure, feed, bedding materials and the animals themselves; ammonia and organic dusts are of prime concern through their effects on pig performance and health (Wathes, 1998). The effects of the aerial environment on pig production were reviewed by De Boer and Morrison in 1988. Their major conclusions still apply today and were that (1) tolerance limits for aerial pollutant exposure have not been defined; (1) potential interactions between aerial pollutants have rarely been examined; (3) dust plays an important part in the aetiology of disease; (4) dusts and gases may reduce productivity directly, or indirectly by affecting health; (5) respiratory diseases are of great economic importance worldwide; and (6) the key features of building design and management to control pollutant exposure are not fully understood.

Much of the early research on lesions induced by exposure to ammonia and dust used concentrations in excess of those found in piggeries and involved exposure for short duration in the absence of specific pathogens (Done, 1991). For example, Drummond *et al.* (1980) reported tracheal and turbinate exudation at 500 ppm ammonia, while Doig and Willoughby (1971) reported tracheal epithelial hyperplasia at 100 ppm ammonia and either 200 mg m^{-3} corn starch or 10 mg m^{-3} corn dust. Conversely, Diekman *et al.* (1993) found no difference in the percentage of lung consolidation and snout grade in gilts exposed to low (4-12 ppm ammonia) or moderate (26-45 ppm) ammonia concentrations. In an epidemiological study, Robertson *et al.* (1990) have found an association between commercial concentrations of aerial pollutants and the incidence and severity of respiratory disease in pigs. Hamilton *et al.* (1996) have shown convincing histological and microbiological evidence of the adverse effects of ammonia exposure in pigs following experimental infection with *P multocida*. Turbinate atrophy was increased significantly at 5 ppm or greater ammonia and was maximised at 10 ppm. They suggest that

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exposure to ammonia facilitates the growth and/or survival of *P. multocida* within the upper respiratory tract, thereby contributing to the severity of the clinical disease of atrophic rhinitis. In a related study, Hamilton *et. al.* (1998) reported an increase in turbinate atrophy with ovalbumin exposure in weaned pigs following *P. multocida* infection, while simultaneous exposure to both ovalbumin (20 mg m^{-3} total mass) and ammonia (50 ppm) caused greater turbinate atrophy than exposure to either pollutant alone (Hamilton *et al.*, 1999).

Guidelines for the tolerance limits of the major aerial pollutants are available but are based mainly upon human exposure CIGR, 1992; Health and Safety Executive, 2001) because of the limited number of studies of the interaction between respiratory disease and aerial pollutants in farm livestock. For example, the recommended limits are 20 ppm for ammonia, and 3.4 and 1.7 mg m^{-3} for the total inhalable and respirable fraction of non specific dusts respectively (Wathes, 1998).

The mechanisms by which poor air quality may interact with respiratory disease and pig performance are complex (Wathes, 1998) but need to be elucidated if our understanding is to be advanced beyond the limitations outlined by De Boer and Morrison (1988).

The principal hypothesis to be tested in a current study is that the incidence and severity of specific respiratory diseases in the weaner pig are greater then combined with chronic exposure to aerial pollutants.

In a recently completed experiment eight groups of 120 weaner pigs, which already had experience of the mixed respiratory diseases of a commercial herd, were be exposed for six weeks to controlled burdens of ammonia and dust. Indicators of disease, such as clinical assessments, post mortem examination and gross pathology, hystopathology and bacteriology of the respiratory tract, and of performance, i.e. growth rate and feed intake, were measured throughout the exposure period to determine the immediate response, as well as post exposure, during the finishing phase of production to test for recovery.

This paper describes the purpose-built facility in which groups of weaned piglets were exposed to controlled concentrations of ammonia and dust. Of particular interest is the ability to generate an aerosol of artificial pig dust, which has not been attempted previously at this scale.

2 General design and construction of the facility

The facility comprises five rooms, each capable of holding 24 piglets, with an access lobby and separate mechanical ventilation and heating systems, a common external slurry pit, an access walkway to the roof ventilation system and an instrument room (figure 1). The unit is based on a proprietary weaner van with additional lobbies and modifications to the ventilation system.

Figure 1:
Facility for controlled exposure to aerial pollutants

The rooms are 2.4 m wide, 3.9 m long and 2.3 m high. Each room is subdivided into two pens, each holding 12 piglets, with a common access. Each pen is equipped with two nipple drinkers and food is supplied *ad libitum* from a plastic hopper. The floor is made of plastic slats. Excreta is collected in a slurry pit, which has sloped floors to a central gully, and is flushed daily with water to minimise the generation of ammonia and other gases. The diluted slurry drains via an air-lock to the common slurry pit. The walls and ceiling are clad with plastic panels without obtrusions, to minimise the area upon which dust can settle.

The husbandry of the animals follows conventional practice. Briefly weaned piglets were drawn from the commercial herd. Lots of 12 piglets were balanced for sex and weight and typically involved litters from 12 sows, i.e. 120 piglets in each batch. The health status of the herd was representative of UK commercial herds. Selected weaner pigs entered the unit at about 3½ weeks of age when they weighed approximately 8 kg and were kept until the end of the experiment at 9½ weeks of age, when they weighed approximately 25 kg. The animals were fed ad-libitum. Detailed assessments of clinical signs of respiratory disease (coughing, sneezing etc.) was made daily and a proportion of the animals were killed at the end of the experiment for pathological, serological and microbiological examination.

3 Environmental control system

Air is supplied to each room under positive pressure by an inlet fan (400 mm diameter, variable speed) and is distributed by a diffuser cone (30^E) placed in the centre of the ceiling. It was exhausted from underneath the slatted floor, to a common exhaust duct, by a single axial fan (630 mm diameter, variable speed). The air inlets were located on the roof of the unit while the discharge of the exhaust duct is approximately 20 m from the unit to minimise cross-contamination. The ventilation rate was identical in all rooms and is set to a constant rate of either 30 or 40 air changes per hour (ach) for winter and summer respectively, after calibration with a Carbon Monoxide tracer using the constant release method (Demmers, 1999). The ventilation rate was not varied in any one trial, except when a period of hot weather was anticipated (defined by an ambient air temperature of 30^oC or hotter) it was then increased temporarily to 60 ach to minimise heat stress. Maintaining a constant ventilation rate allowed a simple system of pollutant generation to be used, without the need for feedback control on the rate of generation of pollutant. However, this required the air to be heated prior to entering the room to maintain the optimum temperature regime.

The manure was removed from the V-shaped pit on a daily basis. The sloping floor was flushed with water, which remained in the pit till the next flushing operation.

The pollutant control system was computer controlled and equipped with a modem for data transmission and remote access. The following parameters were measured: ammonia concentration, ammonia cylinder pressure, ammonia supply flow rate, dust concentration, relative humidity, air temperature, and wind speed and direction. Dust generator motor speed and ammonia supply flow rate were controlled by software been written to run under a Windows operating system. Experimental parameters, such as nominal pollutant concentrations, were selected before each batch began. The software logged all measured parameters and displays the system status including graphs of historical sensor data, the settings and status of the pollutant generators and any warning messages. During exposure, the program turned off the pollutant generator if the measured pollutant concentration exceeded the pre-set safety limits. The software also has separate sections for calibrating and testing the pollutant generators and sensors.

4 Specification, manufacture, generation and monitoring of an artificial pig dust

4.1 Specification of an artificial pig dust

Airborne dust - from any source including a piggery - can be specified in terms of its physical, chemical and microbiological properties (Table 1). While there may be published information on several of these properties, there are few studies in which a full analysis has been undertaken, presumably because of the expense and effort required. Furthermore, since these properties are determined by a host of factors, such as husbandry practices and building design, it is unrealistic to attempt to prescribe the definitive dust for exposure studies. Instead, a test dust may be defined as being representative of the material inhaled by a pig in a piggery if these properties at least are specified and controlled. Clearly there are other properties, such as the particle's electrostatic charge, hygroscopicity and shape (perhaps in terms of the particle size distribution by surface area), that could also be specified but they are probably of lesser importance than those in Table 1 and further research is needed to demonstrate their role in respiratory disease

Table 1:
Important properties of airborne dust

Physical	Concentration of particles by number and mass
	Size distribution of particles by number and mass
Chemical	Chemical composition of toxins
	Chemical composition of allergens
	Source materials
Microbiological	Number of viable bacteria
	Number of non-viable bacteria
	Number of viruses and fungi
	Number of fungal propagules
	Endotoxin content

While there are numerous studies of the mass concentration of dust in pig buildings, there are much fewer in which the size distribution of airborne particles has been measured and even less where the dust composition has been identified by microscopy or any other technique. This is because of the technical difficulties in continuous measurement of particle size and the tedium of microscopic examination. The most comprehensive study is by Heber *et al.* (1988); the principal findings are summarised in Table 2.

Table 2:
Physical characteristics of airborne dust in pig finishing buildings (number of samples=11) (Heber *et al.*, 1988)

	Geometric mean diameter(GMD) µm	Geometric standard deviation (GSD) -	Mass median diameter($MM_{50}D$) µm	Total mass concentration TMC) mg m^{-3}
All particles	2.57 ± 0.33[φ]	2.54 ± 0.13	18.5 ± 3.0	8.1 ± 3.6
Starch particles	12.54 ± 1.13	1.53 ± 0.11	21.0 ± 2.2	-
Grain meal	8.6	1.65	17.9	-

[φ] : Standard deviation

Donham *et al.* (1986) provide complementary information on mass concentration and size distribution, though different sampling techniques were employed. Size distribution was measured microscopically in only one nursery; the geometric mean diameter (GMD) was 2.9 µm with a geometric standard deviation (GSD) of 3.4 µm. This mean diameter is close to the GMD recorded by Heber *et al.* (1988).

In the study by Heber *et al.* (1988), the average total mass concentration was 8.1 ± 3.6 mg m^{-3}. With regard to composition, the feed was found to be the major source of airborne dust particles over 5 µm. This finding is in agreement with Donham *et al.* (1986), Honey and McQuitty (1979) and Curtis *et al.* (1975). Faecal material is the predominant source of fine particles between 1 and 2 µm diameter (Donham *et al.*, 1986), while hair and skin comprised 1 and 10% of particles between 11 to 16 µm (Honey & McQuitty, 1979). Most authors are content to characterise the chemical content of pig dust in terms of the source materials and not specific compounds.

Based on the above studies and taking into account UK production systems, the following specifications of an artificial pig dust were proposed for size distribution: GMD 2.5 µm, GSD 2.5 µm and $MM_{50}D$ 20.0 µm and composition by weight: feed:straw:faecal = 0.5:01:04. The chemical composition was determined by the three sources of material, *i.e*. feed pellets, barley straw and faeces. The faeces were taken from a solid floor of a piggery and contained a small, but unknown quantity of urine. Although skin squames and hair debris are not included specifically in this artificial dust, particles from these sources can be guaranteed in any pig house given the ubiquitous nature of their shedding process.

With regard to the microbial properties, the key question is whether an artificial pig dust should be sterile or not. A sterile dust was specified for mainly pragmatic reasons. First, it is technically difficult to ensure a consistent contamination of the dust with specific microorganisms; and secondly, the release of potentially pathogenic microorganisms in the exhaust air would create a hazard to other pigs on the experimental farm.

4.2 Manufacture of an artificial pig dust

The physical specifications of an artificial pig dust in terms of feed, straw and faeces imply that the dust is manufactured from these ingredients. An equally valid method would have been to collect dust from theair of an existing building, treat it as necessary and then to re-aerosolise it. However, the collection technique could have modified particle size distribution, perhaps by agglomeration and viable microbes deposited on the collection surface could have survived a prolonged exposure to desiccation. Also, the consistency of the dust composition over the duration of the experiment, could not be guaranteed. The technical feasibility of collecting sufficient dust from the exhaust air of one or more buildings was also deemed to be low. Consequently, this method was rejected as unsuitable and an alternative devised that was based on the manufacture of an artificial dust using ‘fresh’ ingredients of known provenance.

Samples of weaner feed, barley straw and faeces (from the floor of other weaner houses) were procured, oven dried at 105°C to constant weight to inactivate microbes and remove moisture and oil binders, milled and mixed. Milling and mixing was carried out by Hosokawa Micron Ltd. (Runcorn, UK) and involved three stages: (1) using a hammer mill (HA 40/32) to pulverise the raw materials into particles of less than approximately 1 mm; (2) using an air classifier mill (15 ACM-SB) to produce a fine dust with a particle size range from 0.1 to approximately 200 µm; and (3) using a mixer (DBX 300) to produce a homogeneous powder. This dust was then triple-bagged and sealed to control the moisture content and prevent contamination before use.

Three lots of pig dust weighing nominally 600 kg in total have been processed. The size distributions of the ingredients from Mix 1 after air milling were measured with a Malvern particle sizer and are given in figure 2.

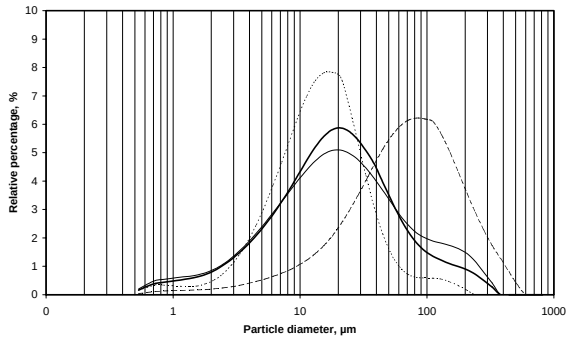

Figure 2:
Relative percentage particle size distribution by volume measured with a Malvern optical particle sizer of three ingredients on an artificial pig dust after milling food (….), barley straw(----), faeces (—)and of the dust after mixing (,,) in the mass ratio 0.5:0.1:0.4 respectively

The number distribution was calculated from the volume distribution assuming spherical particles of unit density, 10^{-6} g m^{-3}. Figure 2 also shows the particle size statistics after the three ingredients had been mixed to form the artificial pig dust. The summary statistics of the volume and number distributions are given in Table 3.

Table 3:
Particle size statistics after hammer milling and air milling.

	Number median diameter ($NM_{50}D$), µm	
Component	Batch 1	Batch 2
Feed	0.68	0.64
Straw	0.73	0.72
Faeces	0.72	0.72
Mix	0.72	0.72
	Volume median diameter ($VM_{50}D$), µm	
Component	Batch 1	Batch 2
Feed	17.7	16.3
Straw	77.3	72.3
Faeces	22.1	19.5
Mix	22.2	23.4

Clearly, air milling created a fine dust of the mix and its constituents. The mix was made by mixing weighed quantities of air-milled components. The volume median diameter ($VM_{50}D$) was satisfactory though the number median diameter ($NM_{50}D$) was lower than the original specification. It may be possible to increase the $NM_{50}D$ by reducing the amount of time for which the components are milled by the hammer or air-classifier mills.

A chemical and microbiological analysis of the dust is shown in Table 4 and is based upon samples taken from mix one and two. Although the micro organism counts were considerably lower, the artificial dust appeared similar in many respects to authentic dust, which was collected from a fully slatted, finisher building. The microbial contamination was low, as expected, because of the thermal inactivation during drying. The endotoxin content was similar to that of 30 µg g^{-1} found by Clark *et al.* (1983). Assuming an inhalable dust concentration of 5 mg m^{-3} and no separation of endotoxin from the dust particles, then this content would gave an airborne concentration of 130 mg m^{-3}, *i.e.* well within the range of 83.5-172.0 mg m^{-3} measured in a recent large-scale survey of pig units (Seedorf *et al.*, 1998).

Table 4:
Chemical and microbiological analysis of artificial and authentic pig dust (fully slatted fattener building)

	Artificial pig dust	Authentic pig dust
Endotoxin content, µg g-1 dust	26.3	32.5
Micro-organisms, Colony forming units g-1 dust		
Total bacteria	5.3 x 103	7.0 x 107
Enterobacteria	0	1.2 x 104
Fungi at 25oC	6.7 x 102	8.1 x 104
Actinomycetes at 32oC	9.2 x 102	3.9 x 104
Actinomycetes at 50oC	3.0 x 102	8.1 x 103
Chemical analysis, %		
Dry matter	95.2	88.1
Crude ash	9.7	19.6
Crude protein	21.3	30.7
Crude fat	4.7	2.8
Crude fibre	10.5	0
N-free extract	49.0	35.1
Trace elements, mg kg-1		
Copper	446	212
Zinc	848	627
Manganese	229	510
Iron	1355	2029

4.3 Generation of an aerosol of artificial pig dust

A number of instruments have been developed over the past 30 years for generation of dust aerosols. Most are used for calibration of particle analysers and no one generator has been developed specifically for long term studies with manufactured pig dust. The principles of operation vary but simplicity and reliability were important criteria when alternative designs were compared. After some development work with Dr W. Koch, Fraunhofer Institut fur Toxikologie and Aerosolforschung, Hannover, Germany, four systems were purchased. The generator comprises an agitated hopper with a horizontal paddle and a high accuracy feeder (GLD 86, FAH, Gericke GmbH, Rielasingen, Germany), equipped with a horizontal auger which meters the dust. Continuous suction from a venturi nozzle, transported the metered dust through the nozzle for delivery into the ventilation system of the weaner unit. The dust clumps were resuspended in the venturi nozzle. This part of the process was extremely reliable. The dust feed rate is controlled by the speed of the feed auger.

The reliability of the generator was poor initially because of stress failure of the horizontal screw auger. The stainless steel spiral auger had been welded around a rod to reduce the dust feed rate but the assembly was too inflexible to accommodate the continuous flexing as the auger rotated. The problem was solved by using a flexible rod only fixed to the motor shaft boss and not attached to the auger. However, occasional failures of the joint between the auger and the boss imply that further work is needed to resolve the problem. The mean feed rate of the dust generated was linearly related to the motor frequency.

4.4 Monitoring the concentration and size distribution of the airborne dust

Dust concentration is measured with three instruments: an AC triboelectric sensor and two gravimetric filter samplers at a single location in each room. The triboelectric sensor (DA50, PCME Ltd) responds to the total electric charge on dust particles and thus provided a continuous estimate of mass concentration. The gravimetric samplers (IOM and SKC cyclone samplers) separate the dust into inhalable and respirable fractions and weekly samples were taken over two day and night periods of 12 h each; the air flow rate is 2.0 l min^{-1}.

Figure 3 shows the continuous measurement of mass concentration over one week at a nominal concentration of 10 mg m^{-3}.

Figure 4 shows the size distribution of dust, as measured with an aerodynamic particle sizer (model 3320, TSI) and the particle size statistics are given in Table 5. The $NM_{50}D$ was higher than that found using the Malvern particle counter (Table 3) for the pure dust before it was dispersed into the room. This is mostly due to the different measurement technique (particle speed versus light scatter) as well as the different nozzles used for the resuspension of the dust. Also the presence of endogenous pig dust from the weaners might have influenced the results. The dust to which the pigs were exposed is finer than originally specified.

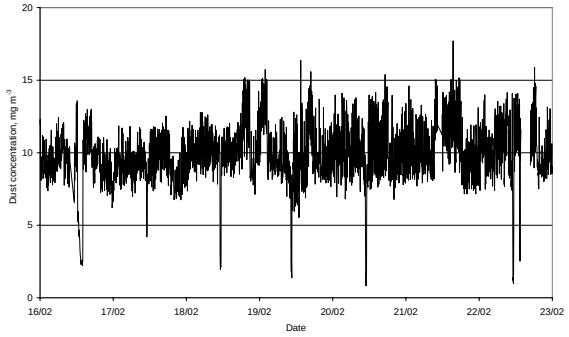

Figure 3:
Dust concentration in room 1 during batch 1 measured using the continuous dust sensor.

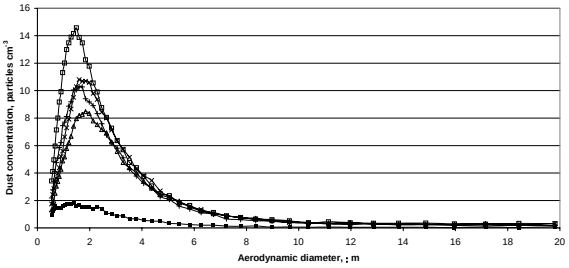

Figure 4:
Particle size distribution at 10 mg m^{-3} on day 35 of exposure; number of samples=4; ♠ room 1, ~ room 2, x room 3, + room 4, □ room5 (control)

The measured dust concentrations over the nine batches of pigs are given in Table 6 and show that the actual average concentrations were within approximately 15% of the nominal concentrations.

Table 5:
Particle size statistics (mean ± sd, n=4) from measurements in the rooms on day 35 of exposure

Room	Number mean diameter, µm		Number median diameter, µm		Geometric mean diameter, µm		Geometric standard deviation -		Mass median diameter, µm	
1	2.44	±0.08	1.86	±0.06	1.93	±0.06	1.90	±0.01	12.7	±0.5
2	2.07	±0.04	1.53	±0.01	1.50	±0.29	1.87	±0.02	13.4	±0.4
3	2.33	±0.05	1.81	±0.03	1.87	±0.03	1.86	±0.02	13.2	±1.0
4	2.16	±0.05	1.66	±0.01	1.74	±0.03	1.86	±0.02	11.8	±1.0
5	1.99	±0.04	1.43	±0.03	1.54	±0.03	1.95	±0.01	14.3	±0.4

The background or natural concentration in the control rooms and in all rooms for batch 2 and 8 was approximately 1.3 mg m^{-3}. Some of the error was due to the equipment's downtime, which occurred when personnel entered the room to tend to the pigs, and during equipment malfunction. This error could have been reduced by incorporating positive feedback in the control for the rate of dust delivery. Furthermore, the rate of generation of endogenous dust from internal sources within the room varied over time, being faster during feeding for example, presumably because dust (from both internal and external sources) settled on building surfaces and was then re-suspended by the activity of the animals. The strength of these sources (1.3 mg m^{-3} could be estimated from the measured concentrations in batch 2 and 8 during which no artificial dust was generated. The relative proportion of artificial and endogenous dust varied according tot he nominal concentration of inhalable dust; this is unavoidable though the absolute quality of endogenous dust was similar between rooms and batches.

Maintaining the lowest dust concentration, i.e. 2.5 mg m^{-3} was difficult, the generators were running at the minimum dust feed rate. During the design phase of the equipment no allowance was made for resuspension of dust. However, due to the unusual climate control system of the facility, the relative humidity was very low (30% - 50%), especially in old winter months. This increased the resuspension of dust. As a result a significantly lower dust feed rate was required than anticipated.

5 Specifications, generation and monitoring of ammonia

The nominal concentrations of ammonia to be maintained in a room were approximately 0, 10, 20 and 40 ppm. Use of a rapid ventilation rate and regular flushing of the slurry pit ensured that the internal sources of ammonia were limited. Ammonia was supplied under pressure from a bottle bank and transported to the inlet duct via fluorinated ethylene polymer (FEP) tubing wrapped with heat trace cable. It was mixed with the inlet air before discharge into each room. The mass flow rate is measured and controlled with a mass flow controller (HITEC F100 Series).

Ammonia concentration was measured with a NO_x chemiluminescent gas analyser after catalytic conversion of NH_3 to NO at 750°C. Gas samples were taken continually from each room at one central location, 1.5 m above the slatted floor, via a switched valve array and vacuum pumps from each room in sequence. The accuracy of measurement of ammonia concentration is 0.1 ppm.

The measured concentration over one week in one room is shown in figure 5 and the performance of the facility for the first nine batches of pigs is given in Table 7. The intermittent falls in ammonia concentration (figure 5) occurred when generation was halted temporarily for staff to service the room. This explains, in part, why the measured average concentrations were lower than the nominal concentrations.

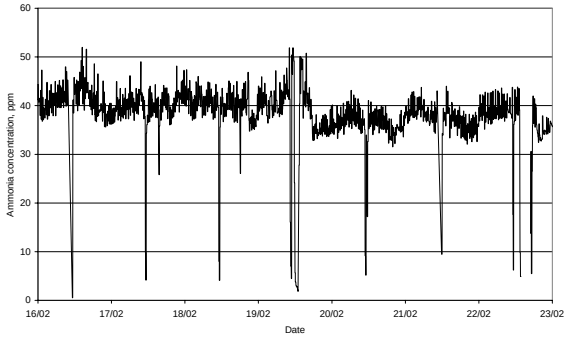

Figure 5:
Ammonia concentration in room 1 during batch 1; spikes are result of ammonia generator down time to allow staff save access to rooms

Table 6:
Mean concentration of inhalable dust (mg m^{-3}) measured in the exposure rooms during the weaner phase

	Mean inhalable dust concentration (mg m^{-3})								
	batch 1	batch 2	batch 3	batch 4	batch 5	batch 6	batch 7	batch 8	batch 9
Nominal	*10*	*0*	*5*	*2.5*	*10*	*2.5*	*5*	*0*	*2.5*
Room 1	11.3 / 12.2	1.5 / 1.3	4.5 / 4.5	2.6 / 2.9	8.8 / 9.4	1.8 / 1.6*	5.2 / 4.9	0.7 / 1.2	1.5 / 1.5*
Room 2	11.5 / 12.8	1.7 / 1.2	1.8 / 1.1*	2.5 / 2.7	1.4 / 1.7*	3.5 / 3.0	0.9 / 1.5*	0.8 / 1.4	2.2 / 2.1
Room 3	11.1 / 10.9	1.8 / 1.6*	5.5 / 4.6	2.6 / 2.8	9.2 / 8.9	5.1 / 5.1	5.0 / 5.3	1.0 / 1.5	2.8 / 3.4
Room 4	9.3 / 11.8	1.5 / 1.4	5.2 / 4.2	2.2 / 2.5	9.0 / 8.9	4.1 / 3.6	4.3 / 4.7	0.6 / 1.0*	3.2 / 3.3
Room 5	2.2 / 2.0*	1.4 / 1.3	5.7 / 6.1	0.7 / 1.5*	8.8 / 9.4	4.6 / 3.6	5.3 / 4.6	0.9 / 1.9	3.3 / 2.9

IOM / continuous sampler; * control room; Batch 6 was abandoned

Table 7:
Mean concentration of ammonia (ppm) measured in the exposure rooms during the weaner phase

	Mean ammonia concentration (ppm)								
	batch 1	batch 2	batch 3	batch 4	batch 5	batch 6	batch 7	batch 8	batch 9
Room 1	0.2	19.8	10.0	19.5	36.8	0.6 *	0.6	0.5	0.8 *
Room 2	9.6	35.2	0.7 *	1.0	1.0 *	0.8	0.6*	17.0	1.0
Room 3	36.0	0.4 *	18.8	11.0	22.0	38.1	10.6	36.2	37.3
Room 4	19.3	9.1	37.3	39.0	0.9	12.3	38.0	0.8 *	9.4
Room 5	0.3 *	0.4	0.0	0.8 *	11.2	21.6	17.3	9.0	17.0

* control room; Batch 6 was abandoned; nominal concentrations 0, 10, 20 and 40 ppm

6 Discussion

The paucity of studies of airborne dust in piggeries has hampered the setting of specifications for dust exposure. After several decades of work on air quality in livestock buildings, little appears to be known about the nature of dust, apart from its mass concentration, which has been measured on numerous occasions. There is a need for the properties of dust, as described in table 1, to be determined. Unless a clear statement can be made of those properties that are important to animal health or environmental pollution, then efforts to control dust generation or emission may well be futile. The deficiency can only be overcome through scientific research leading to clear targets for dust control. For example, the critical component in terms of pig respiratory disease may be the quantity of endotoxin in the respirable fraction of airborne dust. Despite the limited information on dust composition the physical properties of the artificial dust compared well with existing literature values.

The production of artificial dust has been straightforward after the initial problems with the pre-milling preparation were ironed out. Prolonged drying of the feed and faeces to eliminate the emulsifiers was essential to successful milling of these ingredients of the artificial dust.

The new facility has performed satisfactorily. The optional highest ventilation rate of 60 ACH, which was needed to alleviate heat stress, was only required for a short period of two days during batch 7. Standard temperature regimes were maintained throughout the experiment, despite the major changes to the conventional ventilation and heating system. Due to these changes, the relative humidity was lower than would normally be expected in weaner accommodation, which may have affected the respiratory tract of the animals, as well as the survival of micro-organisms in the endogenous dust. Some micro-organisms thrive at low relative humidity, whereas others suffer a detrimental effect. The low humidity could have been avoided if the ventilation rate was not fixed at such a rapid rate. However, this would have complicated the generation of artificial dust, as no reliable online sensor for total dust was available at the start of the project.

The equipment needed to measure and maintain a set concentration of ammonia was straightforward to design and construct and has proved reliable in practice (table 6). In general, the mean concentration in each room was lower than the nominal concentration by 12% or less, due to the periods of downtime of the generator, which were needed for stockman access, and incidental breakdown of the

equipment such as an empty cylinder bank. The high ventilation rate and the daily removal of slurry from the slurry pit successfully reduced the ammonia concentration in the control and zero treatment rooms to 1.0 ppm or lower.

In contrast, the equipment for dust exposure was more complicated for several reasons. Firstly, continuous monitoring of dust concentration is not straightforward. The triboelectric continuous monitors, chosen for their simplicity, required a steady flow of air over the probe provided by an auxiliary fan, shielding from electrical interference (a faraday cage around the probe) and regular cleaning to ensure a reliable measurement of dust concentration. Regular, weekly calibration of the sensor against gravimetric samplers was required. The nozzle of the dust generators proved to be extremely reliable, resuspending the dust consistently into a fine fraction. The weekly measurements with the aerosol particle sizer showed a very consistent size distribution pattern throughout the experiment. However, the feed screw of the hoppers suffered regular mechanical failure and only after redesign of this part was the incidence of failure reduced. Secondly, the resuspension of dust within the rooms by animal activity and the low relative humidity significantly reduced the amount of artificial dust introduced from the dust generators. As a result, it proved difficult to maintain the lower concentration, 2.5 mg m^{-3} (batches 4, 6, and 9) as the generators were required to run at the minimum rate. In contrast to the 5 and 10 mg m^{-3} setting, the mean concentration was invariably higher than 2.5 mg m^{-3}.

The analysis of the production and health assessment data has not been completed to date. The statistical design of the experiment is more sensitive to the possible effects of ammonia over those of dust. Preliminary assessment of the production data suggests that there is no interaction between ammonia and dust exposure. More detailed results, including the health assessment, will be presented at the conference.

7 Conclusions

A facility has been developed in which weaner pigs can be exposed continuously to controlled burdens of aerial pollutants and the effects on health and performance studied at a semi-commercial scale. It has performed satisfactorily throughout the trial. Setting the specifications for dust exposure revealed the many properties of airborne dust and the paucity of information on them. An artificial dust has been successfully produced to the specifications set.

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Livestock related fine dust - composition, structure and flows

H. Takai and S. Pedersen [1]

Abstract

This paper presents an overview of the nature of livestock related fine dust, i.e. composition, structure and flows of dust particles in livestock buildings. Composition deals with contents of chemical components, odorous components, ammonia, bio-aerosols, mites and endotoxin. Structure deals with particle size distributions and effect of porous structure on ammonia desorption from dust particles. Flows deals with aerodynamics of dust particles, spatial dust distribution, dust load on surfaces, particle transport between animals and dust exposure of workers.

Keywords: dust, swine, cattle, poultry, animal building, nature of dust in animal buildings

1. Composition

Dust and airborne dust in animal buildings are a complex mixture of particles of organic and inorganic origin, as well as gases and liquid aerosol absorbed in them. The sources and components of dust are diverse and might include a range of micro-organisms and their cell wall components, dried dung and urine, skin flakes, feathers (in poultry houses), grain mites, spores, pollens, feed and bedding. Dust particles may carry hazards such as pathogenic bacteria, viruses and microbial products and components (such as endotoxin, beta-glucan and peptidoglucans) or other organic substances. Ammonia and odorous components may also be adsorbed on the dust particles.

Chemical Compositions

Aarnink et al. (1999) investigated the composition of dust from different sources in pig and broiler houses, Table 1. For dust in pig houses, the airborne and the settled dust have nearly the same concentration levels of dry matter, ash, N, P, K, Cl and Na. For feed dust, the contents of ash, P and Na are relative low and for faeces the content of Cl is very low. While, skin particles showed high Cl concentration. It was concluded that dust in rooms for rearing pigs mainly originate from feed and skin particles, when the mass is considered.

In comparison with the dust from rearing pig rooms, dust from broiler rooms had lower concentrations of ash, P, Cl and Na, while N and K concentrations were higher in the broiler dust than in the pig dust.

Ammonia in dust particles

The ammonia contents in inhalable and respirable dust sampled in dairy, poultry and farrowing houses were determined by using distillation – indo-phenol method (Takai et al. 1999a). The ammonia contents in dust particles varied from 0.9 to 7.2 µg mg^{-1}.

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Table 1.
Composition of dust from different sources for rearing pigs and broilers.
(Aarnink et al. 1999)

	Dust source	Dry matter g/kg	Ash g/kg	N g/kg	P g/kg	K g/kg	Cl g/kg	Na g/kg
Pig	Airborne dust	921	150	67.0	14.7	27.8	7.83	8.18
	Settled dust	910	120	59.0	11.4	24.4	7.32	6.60
	Feed dust	903	26	21.8	3.4	10.2	7.12	3.58
	Faeces dust	915	149	40.8	20.5	12.7	1.10	3.83
	Skin particles	922	114	67.8	10.7	33.2	15.50	13.00
Broiler	Airborne dust	911	97	169.0	6.44	40.3	4.19	3.23
	Settled dust	914	95	130.0	6.66	29.3	3.46	2.57

Bio-aerosols

Viable bacteria and viruses carried aloft by dust particles may have a greater ability to survive the airborne state (Homes et al. 1996), and cause infections in the barn and in the neighbouring livestock buildings. Dust particles of biological substance (bio-aerosols) show biological action indicated by viability, infectivity, allergenicity, toxicity, or pharmacological activity (Cox and Wathes, 1995). The concentration of airborne micro organisms in animal houses is high. According to Hartung and Seedorf (1999) the incidence of CFU (colony forming units) of bacteria, expressed as log-values, was for pigs 5.1 log CFU/m^3, which is less than for poultry (6.4 log CFU/m^3) and higher than for cattle (4.3 log CFU/m^3). *Enterobacteriaceae* counts for most of the CFU. In the same investigation the mean daily concentration of fungi was 3.7 log CFU/m^3 for pigs, 4.0 log CFU/m^3 for poultry and 3.8 log CFU/m^3 for cattle.

Mites

Mite allergens are thought to affect the respiratory system of humans and animals. In German swine farmers with work-related respiratory symptoms, the concentration of storage mite (type: *Lep d 2*) and house dust mite (type: *Der p 1*, *Der f 1*, *Der2*) allergen in dust, collected from five different sampling sites, was studied in relation to the respective sensitisation rates (Radon et al., 1999). The storage mite allergen (type: *Lep d 2*) was mainly found in the pig confinement house and was abundantly distributed. This allergen was also detected in measurable concentrations in 35 out of 75 farmers' mattresses and in 6 out of 22 urban mattresses. The median concentrations of type *Der p 1* in the farmers' mattresses were significantly higher than in the urban mattresses.

Dust and endotoxin concentration in livestock buildings (Results from an EU-project):

Field surveys of dust and endotoxin concentrations within cattle, pig and poultry buildings were conducted in the United Kingdom, the Netherlands, Denmark and Germany. The study involved about 80 surveys in each country and estimated mean dust and endotoxin concentrations are shown in Table 2 (Takai et al. 1999b).

The overall mean concentrations in cattle buildings were about 0.4 and 0.1 mg/m^3 for inhalable and respirable dust, respectively. Compared with these values, the dust concentrations were 3 to 6 times higher in the pig buildings, and 6 to 10 times higher in the poultry buildings.

Table 2:
Dust and endotoxin concentrations in livestock buildings. (Takai et al. 1999b)

	Inhalable dust Concent-Ration[1] mg/m^3	Inhalable endotoxin		Respirable dust Concent-Ration[1] mg/m^3	Respirable endotoxin	
		Aerial Concent-Ration[2] ng/m^3	Mass Concent-Ration[3] ng/mg		Aerial Concent-Ration[2] ng/m^3	Mass Concent-Ration[3] ng/mg
Cattle building						
England	0.22	-	-	0.15	-	-
The Netherlands	0.30	16	53	0.09	2	24
Denmark	0.39	13	33	0.04	1	13
Germany	0.65	14	21	0.05	0	4
Overall mean	0.38	14	38	0.07	1	13
Pig building						
England	1.87	54	29	0.24	4	16
The Netherlands	2.43	118	49	0.25	10	39
Denmark	2.76	95	34	0.26	11	42
Germany	1.95	33	17	0.18	9	52
Overall mean	2.19	67	31	0.23	7	31
Poultry						
England	3.31	603	182	0.51	52	101
The Netherlands	4.58	132	29	0.58	11	19
Denmark	4.52	102	23	0.64	22	35
Germany	2.22	128	58	0.19	10	54
Overall mean	3.60	200	56	0.45	21	46

1) Takai et al., 1998. 2) Calculated on basis of Seedorf et al., 1998-C.
3) (Endotoxin concentration)/(Dust concentration)

The overall mean values for inhalable and respirable aerial endotoxin concentrations in cattle buildings were 14 and 1 ng/m^3, respectively. Compared with these values, there were 5 to 7 times higher values for pig buildings, and 14 to 21 times higher values for the poultry buildings.

While, looking at the mass endotoxin concentrations, i.e. the endotoxin concentration expressed in terms of ng of endotoxin per mg of dust, the type of animals showed less effect. The endotoxin mass concentrations in inhalable dust were about the same level for all three animal species. The endotoxin mass concentrations in the respirable dust in the pig and the poultry buildings were generally higher than the concentrations in the cattle buildings. In the cattle buildings, the mass endotoxin concentrations in the inhalable dust were about 3 times higher than the concentrations in the respirable dust.

If the aerial endotoxin concentration must be lower than 10 ng/ m^3 regarding to the occupational exposure limit, the inhalable dust concentration should be lower than 0.3 mg m^{-3}. This will be very difficult to achieve in livestock buildings, especially in pig and poultry buildings.

Odour and dust

Dusts in livestock ventilation air have been implicated in transporting and even magnifying odour. Mechanisms suggested as being associated with this include odorous molecules (odorants) being absorbed on particle surfaces and then desorbing in large local concentrations in the nasal lining, where the olfactory nerve cells are located (Williams, 1989; Hammond et al. 1981). Dust in pig houses contains volatile fatty acids (*VFA*) (Hartung, 1985; Williams, 1989), which are assumed to be the indicator compounds for the odour. However, the relationships between dust concentration and odour intensity is not clear (O'Neill and Phillips, 1992). Further research on absorption and desorption of odorants on dust particle surfaces is needed to understand the relationship between odour and dust.

2. Structure

The number median aerodynamic diameters of dust from animal houses are ca. 1.5 - 2 µm with standard deviation of about the same values. Figure 1 shows an example for particle size distribution of

swine dust. Figure 2- 4 shows electron microscopic pictures of dusts sampled in houses for dairy cows, layers and pigs, respectively. The observation of dust particles showed that the particles from the dairy house have relatively smooth surfaces, and the particles from the poultry house have rough surfaces. The surface structure of the dust particles from the farrowing house was somewhat between the other two ones.

The porous structures of dust particles may restrict desorption of gas molecules, and allow slow and continuous gas emissions from dust particles over a long period. Takai et al. (1999a) demonstrated this by following experiment: About 3g of the sediment dust was packed between two glass fibre filters in a 37 mm dust sampler cassette, and exposed to clean air for 48 hours with a air flow rate of 1 l m^{-1}. Theammonia emitted to the air was determined by using 0.5% boric acid trap and indo-phenol method.

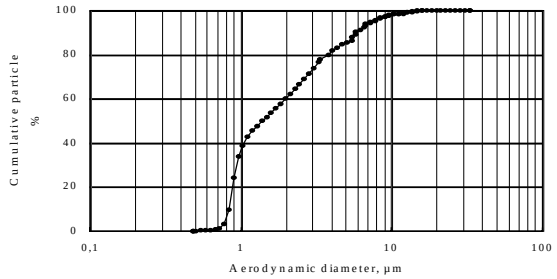

Figure 1:
Example for particle size distribution of swine dust

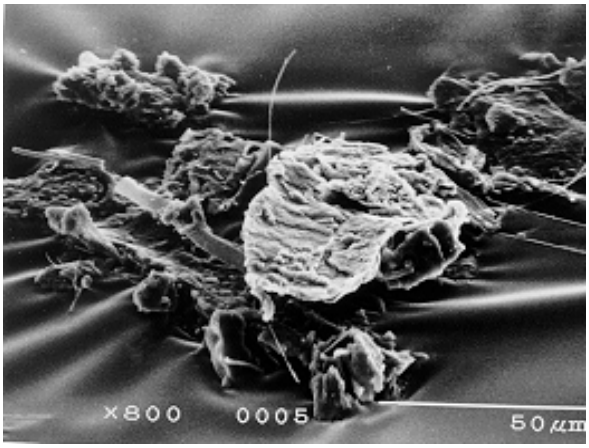

Figure 2:
Electron microscopic view of airborne dust collected in a house for dairy cows.

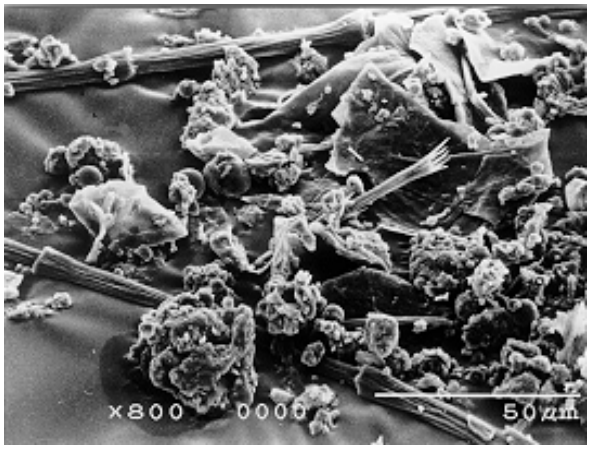

Figure 3:
Electron microscopic view of airborne dust collected in a free-range system for layers.

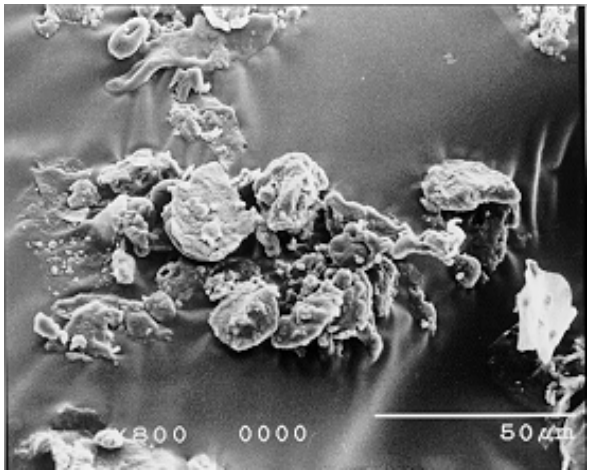

Figure 4:
Electron microscopic view of airborne dust collected in a farrowing house.

The rate of ammonia emission, µg NH_3 s^{-1} g^{-1}, decreased quickly in the first exposure hours to a level, where the rate of ammonia emission decreased slowly over a long exposure period. It was from about 0.02 to 0.004 µg NH_3 s^{-1} g^{-1} during an exposure period from 4 to 48 hours. This indicates that the ammonia molecules are bound in a dust particle at different energy levels. The ammonia molecules, which were emitted in the first exposure hours, were bound at law energy level, i.e. on the surface, while the remainder was bound at a higher energy level, i.e. in the porous structure and/or absorbed in the body of the particles.

3. Flows

Forces affecting on particles in air

Particles in air are affected by a variety of forces (e.g. aerodynamic, gravity, buoyancy) and undergo turbulent coagulation, turbulent diffusive deposition, and gravitational sedimentation (Andersen, 1995; Liao and Feddes, 1991). Larger particles, say larger than 5 µm, deviate in their movement significantly from the air stream. A particle near the surface or near the other particles is affected by van der Waals forces. Under a high dust concentration, as in animal houses, the behaviour of small particles may be affected by particles with greater mass.

Spatial dust distribution

Dust is not uniformly distributed within an animal building, and its behaviour depends on air distribution, relative locations to the dust sources, animal and human activity levels in the buildings and air cleaning technologies (Barber et al., 1991; Smith et al., 1993; Takai et al., 1996; Wang et al., 1999). The overall dust levels during the day are generally higher than during the night, due to animal activity that increases the airborne dust concentration, as shown in figure 5 (Pedersen and Takai, 1999). Ventilation rate have a large effect on the dust spatial distribution. Increasing the ventilation rate reduces the mean dust concentration in the room until some ventilation rate, above which the ventilation rate has less effect on dust concentration. Wang et al. (2000) showed that dust mass concentration varied as much as 30-fold between the lowest and the highest within a mechanically ventilated room.

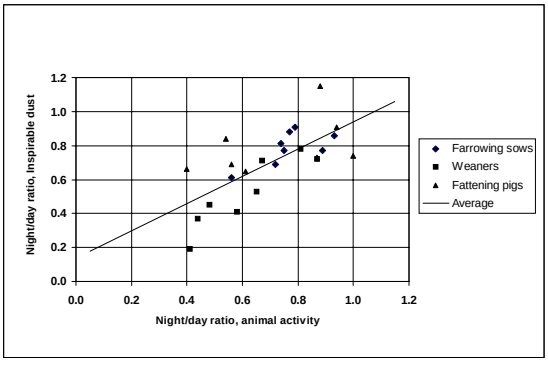

Figure 5:
Correlation between animal activity and inspirable dust concentrations in pig houses

Ventilation rate affects relative humidity and moisture level of the surfaces in the animal building. Increased ventilation rate may, therefore, result in dryer surface and increased dust development from the surfaces due to animal activity. In such case, the dust concentration will increase with increasing ventilation rate, which is may be caused by increased animal activity.

Dust load on surfaces

Dust load is the difference between deposited and resuspended dust. The dust load is highest on the floor but dust loads on the walls and the ceiling occur. The measurements carried out by Lengweiler (2000) indicate that the air velocity has a non-linear influence and that the turbulence has a larger effect on the deposition than on the resuspension. Therefore high turbulence causes high dust load. However, the influence of turbulence and velocity are strongly dependent on each other and cannot be analysed in isolation. Langeweiler (2000) has measured deposition velocity of talcum powder on floor, wall and ceiling with different turbulent kinetic energies, and developed linier regression models as shown in Table 3.

Table 3:
Linear regression models for deposition velocity, a, as function of turbulent kinetic energies, k [$m^2 s^{-2}$] (Lengweiler, 2000)

	Deposition velocity a [$m s^{-1}$]	Standard error S [$m s^{-1}$]	r
Floor	$1.41\times10^{-3}k + 6.36\times10^{-4}$	2.0×10^{-4}	0.82
Wall	$2.79\times10^{-4}k + 1.94\times10^{-5}$	2.1×10^{-5}	0.94
Ceiling	$7.56\times10^{-5}k + 1.33\times10^{-5}$	5.7×10^{-6}	0.94

Particle transport between animal in a room

To obtain an improved understanding on transport and spreading of pathogenic bacteria in animal houses, physical mechanism behind the airborne particle transport between animals has been studied (Takai et al., 2000). The experiments have been carried out in a full-scale test room by using so-called a cough simulator. It showed that the mean resultant air velocity, turbulence intensity and time of particle transport affect significantly on how big part of particle released from one animal can reach other animals. On the other hand, turbulence time scale, length scale and diffusivity did not show significant effect.

Dust exposure of workers in livestock buildings

Dust exposures reported in literature show very wide range, from less than 1 mg m^{-3} to more than 40 mg m^{-3} of total dust (Vinzents and H. Christensen, 1990; Takai et al., 1996). Workers in livestock building perform many different types of work, and dust exposure is highly depending on the kind of work. How the work is performed affects also dust exposure. For example, handling of feed in ruffled manner may generate more airborne dust than when it is handled in unruffled manner. Vinzents and Christensen (1990) reported that about 40% of the working time in pig buildings involved close contact with pigs, e.g. weighing of pigs. During this type of work, farmers are exposed to high concentrations of dust caused by increased animal activity, which disperse dust from animals and surfaces of the building. Furthermore, the work is often strenuous so that the farmers will inhale large amounts of dust.

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Comparison of the indoor dust concentration between two different housing and ventilation systems for fattening pigs

Eva Gallmann, E. Hartung and T. Jungbluth [1]

Zusammenfassung

Vergleich der Innenraum-Staubkonzentration unterschiedlicher Haltungs- und Lüftungssysteme für Mastschweine

Gegenstand der Untersuchungen war ein Vergleich des Tagesverlaufs und der durchschnittlichen Staubmassenkonzentration (PM10, PM 2,5, PM 1,0) im Innenraum zwischen einem Vollspaltenbodenstall mit Zwangslüftung (FSF) und einem Haltungssystem mit getrennten Klimabereichen und freier Lüftung (KN) für Mastschweine. Die Messungen wurden jeweils an drei bis vier Tagen von vier aufeinanderfolgenden Mastdurchgängen zwischen Herbst 1999 bis Frühjahr 2001 durchgeführt. Der Haupteinflussfaktor auf den Tagesverlauf der Staubkonzentration war die Tieraktivität, die wiederum maßgeblich vom Fütterungsregime bestimmt wurde. Die mittlere PM 10 Innenraumstaubkonzentration aller Messungen betrug 0,46 mg/m³ im FSF System und 0,17 mg/m³ im KN System. Ebenso waren die durchschnittliche PM 2,5 und PM 1,0 Staubkonzentration im System FSF mit 0,15 mg/m³ bzw. 0,12 mg/m³ höher als im System KN mit 0,07 mg/m³ bzw. 0,06 mg/m³. Die Unterschiede zwischen den Haltungssystemen hinsichtlich der Tagesdurchschnittswerte der Staubkonzentration, Innenraumtemperatur und relativer Luftfeuchte sowie des Volumenstroms waren signifikant im Gegensatz zur Stallbelegung in Großvieheinheiten. Als Hauptgründe für die durchweg geringeren Staubkonzentrationen im Innenraum des Haltungssystems KN werden die höhere Luftfeuchte, der größere Anteil feuchter Flächen und im Mittel höheren Volumenströme angesehen. Innerhalb des jeweiligen Haltungssystems konnte ein signifikanter negative Korrelationsfaktor zwischen der Staubkonzentration und dem Volumenstrom bezogen auf die Großvieheinheit bestimmt werden. Eine Berechnung von PM Emissionen aus den dargestellten Ergebnissen zur Innenraumstaubkonzentration ist nicht zulässig.

Schlüsselworte: Staub, PM 10; 2,5; 1,0, Mastschweine, Stallluftqualität, Tieraktivität, Lüftung

Abstract

In this research work the diurnal course and mean indoor dust mass concentration (PM 10, PM 2,5 and PM 1,0) of a housing system for fattening pigs with fully slatted floor, forced ventilation (FSF) and a kennel housing system with natural ventilation (KN) were compared. The experiments were carried out during three to four measuring days of four subsequent fattening periods in the time between autumn 1999 and spring 2001.

The main influence on the diurnal course was the animal activity, which itself mainly was determined by the feeding strategy. On average the PM 10 indoor dust mass concentration of all measurements was 0,46 mg/m³ for the FSF housing system and 0,17 mg/m³ for the KN housing system. In the same way higher values for the medium PM 2,5 indoor concentrations were found for the FSF system with 0,15 mg/m³ in contrast to the KN system with 0,07 mg/m³. Equivalent results were achieved comparing the average PM 1,0 indoor dust mass concentration, which came to 0,12 mg/m³ for the FSF system and 0,06 mg/m³ for the KN system respectively. The daily mean values of the dust concentration, indoor temperature, relative humidity and airflow rate between the two housing systems differed on the whole significantly (P=0,01), whereas the slight differences in livestock units could be neglected.

The higher air humidity, higher share of damp surfaces and on average higher airflow rates are considered to be the main reasons for the over all clear lower indoor dust mass concentration in the KN housing system. Within each housing system a significant negative correlation factor between the mean PM 10 dust concentration and air flow rate per livestock unit could be determined. Because of very likely alterations of the particle characteristics, mass and number concentration in the exhaust air compared to the indoor air, a calculation of PM emissions from the described results of the indoor dust concentrations is not permissible.

Keywords: Dust, PM 10; 2,5; 1,0; fattening pigs, indoor air quality, animal activity, ventilation

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1 Introduction

Dust concentrations or so called airborne particles respectively its containing potentially harming compounds undoubtedly worsen the indoor air quality and contribute to respiratory health problems in both animals and people.

Organic dusts in livestock buildings comprise grain and other plant-derived particles, animal hair, skin, urine, faeces, microorganisms and other particles. Dust particles may carry hazardous material such as pathogenic bacteria, viruses, endotoxin or other organic substances. Viable bacteria and viruses carried in the air by dust particles may have greater ability to survive, and cause infection in the animal house and in neighboring livestock buildings. Dust particles of biological substances (bioaerosols) show biological action indicated by viability, infectivity, allergenicity, toxicity or pharmacological activity (Takai et al., 1998).

Further more questions concerning the environmental impact and feared adverse health effects of bioaerosol emissions and immissions become increasingly focus of public and political interest. For some years the emission and transport of fine particulate matter (PM) has been considered by international strategies of air pollution and control. Recently, the agricultural panel of the UNECE task Force for Emissions Inventory and projections is dealing with PM 10 and PM 2,5. Particulate matter becomes explicitly subject to national and European regulations regarding agricultural sources and livestock husbandry. These efforts are among others currently reflected in the definitions of the “Best Available Techniques (BAT)”in the frame of the IVU-guideline (EU Directive 96/61/EG, 24.09.1996) (UBA, 2001) or in the German draft regulation for minimum distances between livestock buildings and residential areas (BMU, 2001). The quality of emission inventories and derived avoidance strategies however, are mainly dependent upon the availability and the quality of data regarding source specific emissions.

Therefore the investigation of factors influencing dust concentration and composition in pig houses is one important topic in order to develop dust control mechanisms. The examination of the dust pollution is consequently an essential part of comparative studies to evaluate different pig housing systems.

2 Objectives

In this research work special emphasis was laid on the comparison of the indoor dust concentration between a forced and naturally ventilated pig housing system for fatteners. On the one hand it was focused on the interactions between dust concentration, animal activity and feeding strategy. On the other hand attention was paid to monitor the amount, range, and dynamics of the indoor dust concentration in the course of four subsequent fattening periods from autumn 1999 till spring 2001. The dust measurements were one aspect of investigations dealing with an extensive evaluation of two different housing and ventilation systems with regard to animal behaviour, indoor air quality and the environmental impact of odour and gaseous emissions (Bea et al., 2001; Brose et al., 2001; Gallmann et al., 2001; Gallmann et al., 2002; Martinec et al., 2000; Schneider et al., 2001).

3 Approach

3.1 Pig housing system

The comparative studies between the housing systems “fully slatted floor, forced ventilation” (FSF) and “kennel housing, natural ventilation” (KN) were carried out in two spatially separated compartments of the experimental pig facility for fattening pigs in Hohenheim. Figure 1 shows the ground plan of the pig house with the arrangement of the sampling points and measured parameters.

The system FSF was divided into 6 pens (fully slatted floor) with 9 animals each and the forced ventilation system was designed as underfloor extraction with fresh air supply though pore channels. The system KN featured 2 pens with 24 animals each divided into an activity area with slatted floor and a covered, level concrete resting area per pen. The natural ventilation in the KN housing system was designed as a gravity shaft system with an additional solar-powered fan in the eastern shaft for optional supporting ventilation. Fresh air was admitted to the compartment along the western side (mean wind direction) of the building.

Each pen was additionally equipped with a pig occupation technique (“Porky Play”) using straw, chains and wooden beam (Stubbe and Jungbluth, 1999). Both housing systems featured the same feeding strategy using a sensor liquid feeding system with 16 feeding times per day from 6:00 a.m. to 10:00 p.m.

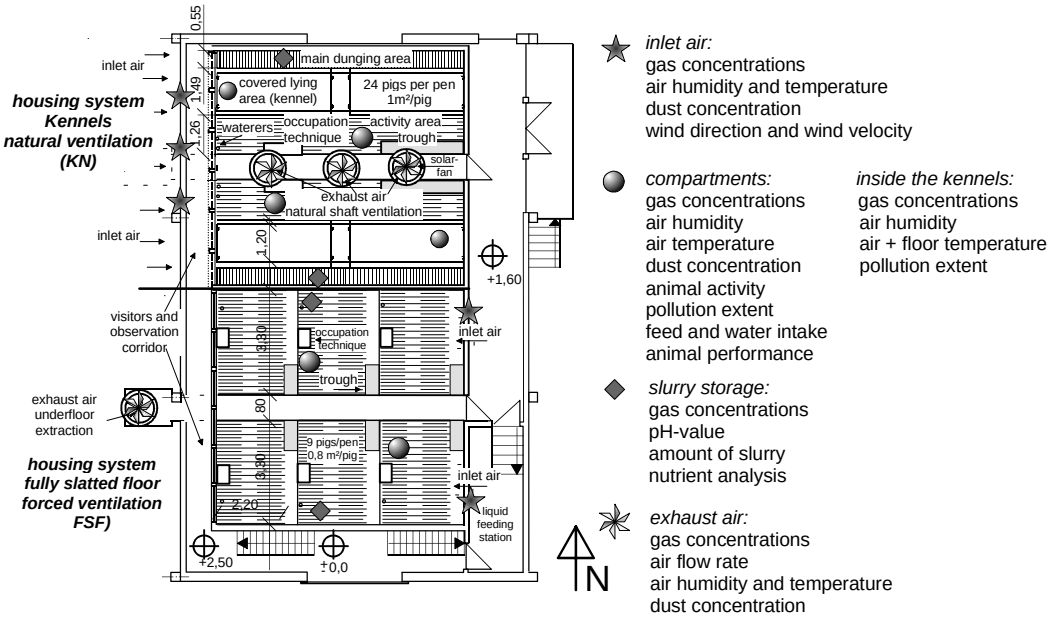

Figure 1:
Ground plan of the experimental pig facility and housing systems with arrangement of the sampling points and measuring parameters

For demanuring, the liquid manure was collected in both systems in two containers each on both sides of the feed passage over the entire fattening period and drained only after the animals had been stalled out.

The compartments were stocked at the same time according to the all in – all out principle with a fattening period normally running from 25 kg till 110 kg life weight, lasting approximately 105 days.

3.2 Dust measurement

For the determination of the dust mass concentration six aerosol monitors (90 degree light scattering laser photometer) "Dusttrack ™" (TSI) were in use. The sensitivity of the devices is characterized by the manufacturer as follows: Particle mass range is 0,001 – 100 mg/m³ with a resolution of ±0,001 mg/m³, the size range is 0,1 - ~10µm. The flow rate was adjusted to 1,7 l/min.

Light scattering-type aerosol monitors respond linearly to the aerosol mass concentration. The scattered light is dependent upon particle size. This dependence is most dramatic for particles with diameters less than one third the wavelength of the laser (~0,25 µm). The scattered light is also dependant upon the index of refraction and light absorbing characteristics of the particles. The internal calibration factor for calculating the dust mass concentration can be adjusted to the specific aerosol, when a simultaneous gravimetric analysis is carried out.

By the use of the corresponding inlet conditioner the upper particle size could be limited for each device at 10, 2,5 or 1,0 µm. According to the ISO standard, the critical particle diameters corresponding to 50 % sampling efficiency (D50) are defined as follows. The PM 10 (particulate matter with a diameter < 10 µm) so called thoracic fraction is that portion of the inhalable (PM 100) particles that pass the larynx and penetrate into the conducting airways and the bronchial region of the lung. The PM 2,5 and PM 1,0 fractions are part of the respirable fraction (PM 4) that enters the deepest part of the lung, the non-ciliated alveoli.

Dust measurements were carried out up to four times during each fattening period for at least 48 to 72 hours with a scanning rate of 30 seconds. The aerosol monitors were run parallel and placed always in the same pen of each housing system near the feeding passage and the trough at comparable places. It follows that at the KN housing system only a sampling of the indoor air of the activity area (c.f. figure 1, section 3.1) but not inside the kennels was carried out.

The height of the sampling point was about 1 m above the floor. The aerosol monitors used for the different measurements were in each case randomly selected from the pool of the six devices.

The zero point of the aerosol monitors was controlled before and after each measurement. Values which were shown to be incorrect and illogical in a plausibility check were rejected without any data

interpolation. For the main purpose of monitoring and comparing the course and amount of the indoor dust concentrations of two different pig housing systems, the "Dusttrack™" aerosol monitors have been proved to be very handy, robust and satisfactory accurate. With the help of additional parallel measurements with an more precise optical particle counter "Dustcheck™" (Grimm Inc. model 1.108) (Schneider et al., 2001) the performance and accuracy of the six aerosol monitors was regularly checked. The optical particle counter classifies the particle number concentration into 15 size channels between 0,3 and 20 µm diameter (optical latex equivalent diameter) and offers the opportunity to calibrate the according mass concentration.

Further surrounding parameters such as animal activity, airflow rate (measuring fan), air temperature and humidity were registered continuously over the entire fattening period (Gallmann et al., 2000). The animal activity was recorded with the same scanning rate as for the dust concentration of 30 seconds with two PID sensors (Passiv Infrared Detector) (Pedersen and Takai, 1999) in each compartment.

For the results of the daily mean dust concentrations presented in this paper, always the values of the second measuring day from 0:00 a.m. to 0:00 p.m. have been considered. The daily mean of 24 hours is therefore based on 2880 individual values.

4 Results and Discussion

4.1 Diurnal pattern of dust and animal activity

The diurnal variation of the indoor dust concentrations showed in all cases a clear parallel course with the animal activity signal. Figure 2 demonstrates with the help of an example of the first day of a fattening period inside the FSF housing system the development of both parameters before and after the pigs have been stalled in. The sensitivity of both parameters is mirrored by the reaction on the handling of straw for filling the occupation technique before the pigs were stalled in.

Similar effects were found by Hinz et al. (1999) during putting straw into a cattle house. Because of the irregular action of animals and workers during the preparation and filling of the compartment from 12:00 to 18:00, both parameters do not have a clear parallel course as it is observed in the evening hours. In the evening the animals calmed down, thus the main influence factor on the activity and following increase in dust concentration were the feeding times.

Typical is the lower level and range of both parameters during night time, whereas of course likewise movements of the animals even at night time occur and are mirrored in the activity and dust signal.

The striking conformity between the animal activity and dust signal was also found at the same pig house by Schneider et al. (2001) when using an optical particle counter (Dustchek™, Grimm) (c.f. section 3.2). Thus the animal activity induced particle release and so increase of the particle number both in micron and submicron size ranges. This is above all important regarding the higher health risk of small particles. Gustaffson (1999a) found a similar daily variation in number of dust particles of different sizes in connection with the animal activity.

The animal activity sensor is all in all a very good, handy, cheap and robust indicator from which a diurnal course of the indoor dust concentration can be derived. It offers the opportunity to reduce the dust measurement itself to carefully selected measuring periods.

The main influence exerting on the animal activity again was the particular feeding strategy in both housing systems with 16 feeding times between 6.00 a.m. and 10:00 p.m.. In figure 3 the mean diurnal pattern of the indoor PM 10 dust mass concentration of the FSF and KN housing system is compared. The mean diurnal pattern was calculated from 13 (FSF) respectively 14 (KN) 24 hours measurements between October 1999 till March 2001 during different seasons and fattening stages (c.f. Tab 1, section 4.2).

During feeding short peaks of the mean dust mass concentration were at least about twice up to six times the amount between the feeding times. The increase of the dust mass concentration during the feeding happened very fast within a few minutes, whereas the decrease after the feeding needed longer up to half an hour, presumably until the pigs have calmed down again. Even if there is at both housing systems a similar mean diurnal pattern in dependence of the feeding times, the dust mass concentration between the feeding times as well as of the peaks during feeding is remarkable lower for the KN housing system (c.f. section 4.2).

The strong influence of the animal activity and feeding strategy on the indoor dust concentration has been proven by several authors (in Gustafsson, 1999a).

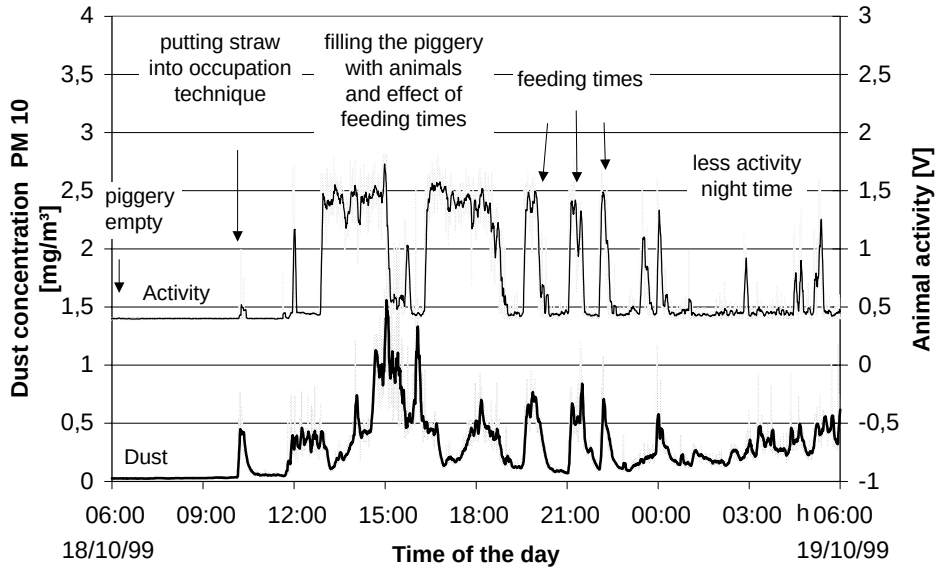

Figure 2:
Comparison of the course of the activity signal and PM10 dust concentration during the first day of one fattening period

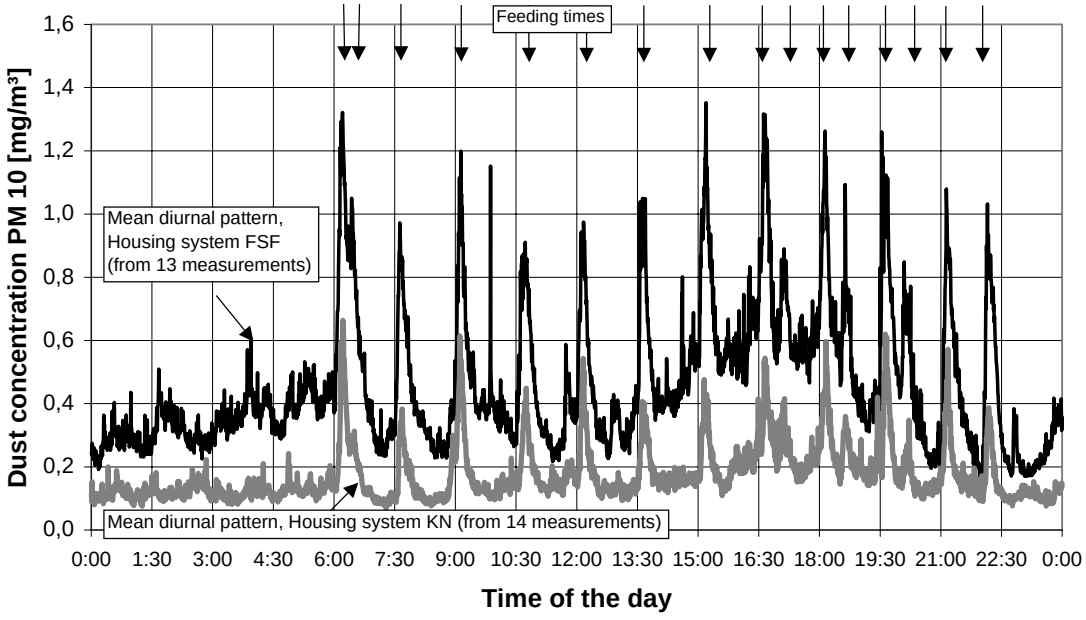

Figure 3:
Comparison of the mean diurnal pattern of the indoor dust concentration (PM 10) between the housing systems for fattening pigs fully slatted floor, forced ventilation (FSF) and kennel housing, natural ventilation (KN), calculated from 13 (FSF) resp. 14 (KN) measurements in the time from autumn 1999 to spring 2001

4.2 Comparison of the indoor dust concentration

The comparison of the mean diurnal pattern of the indoor dust concentrations (figure 3) already pointed to the lower mean and range of the dust mass concentration determined for the KN housing system. Figure 4 and 5 underline by means of plotting the results as box-plots[2] the observation, that during all measurements clear differences regarding the mean, median and variation of the measured PM 10 values existed. Especially the maximum values measured in the FSF housing system were often two to five times higher than in the KN housing system. The same assessment applies to the results of the PM 2,5 and PM 1,0 comparative measurements in both housing systems.

Table 1 comprises a detailed survey of all results of the dust mass concentrations in connection with the respective daily mean values of the indoor temperature, indoor relative humidity and airflow rate. On average the PM 10 indoor dust mass concentration of all measurements was 0,46 (0,17-0,91) mg/m³ for the FSF housing system and 0,17 (0,12-0,25) mg/m³ for the KN housing system. In the same way higher values for the medium PM 2,5 indoor concentrations were found for the FSF system with 0,15 (0,09-0,22) mg/m³ in contrast to the KN system with 0,07 (0,03-0,11) mg/m³. Equivalent results were achieved comparing the average PM 1,0 indoor dust mass concentration, which came to 0,12 (0,08-0,13) mg/m³ for the FSF system and 0,06 (0,05-0,07) mg/m³ for the KN system respectively. The daily mean values of the dust concentration, indoor temperature, relative humidity and airflow rate as given in table 1 between the two housing systems differed on general significantly (P=0,01), whereas the slight differences in livestock units can be neglected.

Additionally, the correlation by Pearson between the PM 10 daily mean dust concentration and different influencing factors within each housing system was tested. For the FSF housing system a significant negative (P = 0,001) correlation coefficient (r = -0,789) of the PM 10 dust concentration only with the air flow rate per livestock unit [m^3*h*LU^{-1}] could be determined. A similar but weaker relationship between the air flow rate per livestock unit and PM 10 dust concentration was also found for the KN housing system, with a correlation coefficient by Pearson of r = –0,589 and significance level of P = 0,05. The corresponding linear regression factors were $r^2 = 0,622$ (FSF) and $r^2 = 0,347$ (KN). That is to say that only around 62 % (FSF) and 35 % (KN) of the variation of the mean dust mass concentration can be explained by the influence of the air flow rate per livestock unit.

This results hint at the effect, that to a certain extend (besides the interaction of several influencing factors) the mean dust concentration decreases, when the air flow rate related to the livestock unit increases or the other way round a decrease of the air flow rate per livestock unit results in an increase of the mean indoor dust concentration.

The reference to the livestock unit implies of course the effect of the fattening day respectively weight gain of the pigs. The airflow rate on the other hand is mainly influenced by the temperature control of the forced ventilation of the FSF housing system, whereas the natural ventilation of the KN housing system mostly depends on the driving forces for the air exchange which are wind pressure, temperature difference and the optional use of the solar fan.

Concluding from the described results the higher air humidity, higher share of damp surfaces and on average higher airflow rates are considered to be the main reasons for the over all clear lower indoor dust mass concentration in the KN housing system.

Although an additional explanation can be seen because of differences in the air distribution inside the two housing systems along with a known variableness of the spatial distribution of the dust concentration inside a room (Wang et al., 2000; Hinz and Linke, 1998). For the kennel housing has to be taken into account that large differences in dust concentration inside and outside the kennel are according to experiments of Mayer and Hauser (1999) to be expected, mostly depending on the animal activity and presence of the pigs inside or outside the kennel.

Similar effects were found e.g. by Gustaffson (1999b) who determined significant lower dust values for a cold confinement with straw bedding and natural ventilation compared with an insulated climate controlled barn in total and respiratory dust concentration and settling of dust. A possible explanation for the large differences might have been the higher moisture in the cold environment with straw bedding.

[2] Boxplot: The horizontal lines in the box denote the 25^{th}, 50^{th} and 75^{th} percentile values. The error bars denote the 5^{th} and 95^{th} percentile values. The two symbols below the 5^{th} and above the 95^{th} percentile error bar denote the 0^{th} and 1^{st} respectively the 99^{th} and 100^{th} percentiles. The square symbol in the box denotes the mean of the measured of data.

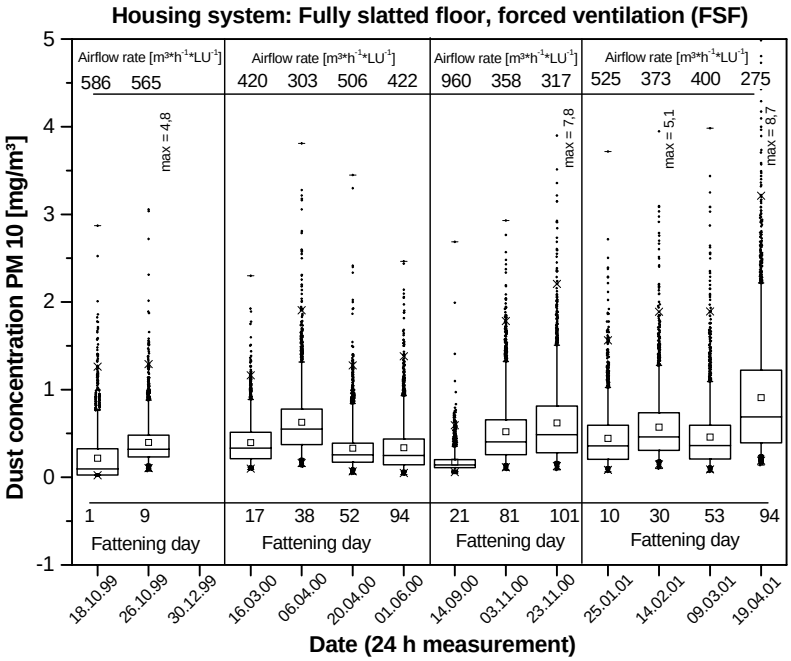

Figure 4:
Indoor dust concentration (PM 10) of several 24 hours measurements during different fattening periods at the housing system fully slatted floor, forced ventilation (FSF) (LU = 500 kg liveweight)

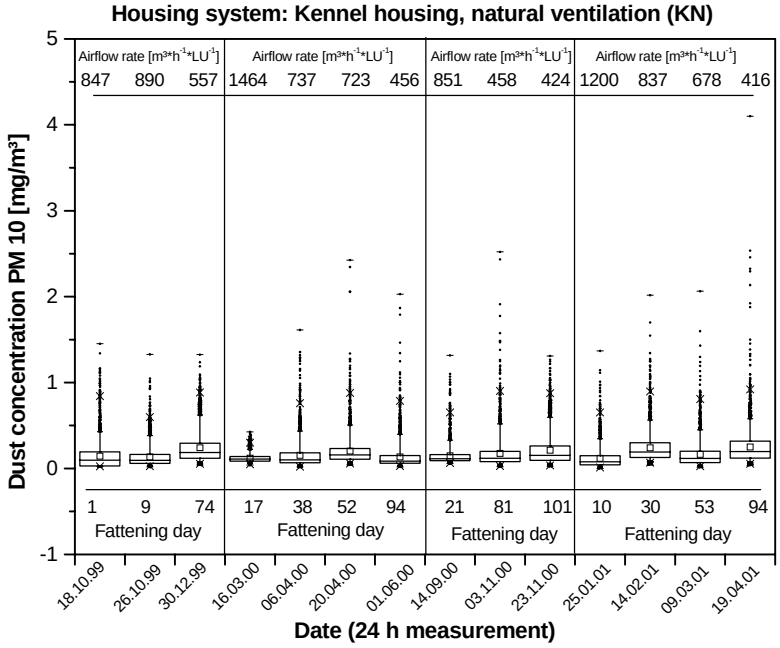

Figure 5:
Indoor dust concentration (PM 10) of several 24 hours measurements during different fattening periods at the housing system kennel housing, natural ventilation (KN) (LU = 500 kg liveweight)

Table 1:
Summarized results of the comparison of the indoor dust concentrations and surrounding parameters between two housing systems for fattening pigs

Date, Fattening day	Housing system, (Livestock Units = LU)	Particle size < [µm] (PM)	Daily mean, (minimum-maximum) dust concentration	Daily mean values		
				Indoor Temperature [°C]	Indoor rel. humidity [%]	Air flow rate [$m^3*h^{-1}*$ pig^{-1}] / [$m^3*h^{-1}*LU^{-1}$]
18.10.99 Day 1	FSF (3,5)	10	0,22 (0,02-2,87)	19	42	38 / *586*
		2,5	0,09 (0,02-0,73)			
		1,0	0,13 (0,03-1,86)			
	KN (3,4)	10	0,14 (0,02-1,45)	11	52	60 / *847*
		2,5	0,06 (0,02-0,48)			
		1,0	0,07 (0,02-0,45)			
26.10.99 Day 9	FSF (4,3)	10	0,40 (0,07-4,84)	22	57	45 / *565*
		2,5	0,10 (0,03-1,44)			
	KN (4,1)	10	0,14 (0,02-1,33)	16	70	76 / *890*
		2,5	0,04 (0,01-0,25)			
30.12.99 Day 74	FSF	---	---	---	---	---
	KN (8,7)	10	0,24 (0,04-1,33)	5	78	101 / *557*
16.03.00 Day 17	FSF (4,5)	10	0,40 (0,09-2,30)	18	49	35 / *420*
	KN (4,0)	10	0,12 (0,04-0,43)	6	86	122 / *1464*
06.04.00 Day 38	FSF (6,6)	10	0,63 (0,12-3,81)	20	52	37 / *303*
		2,5	0,18 (0,03-1,37)			
	KN (5,8)	10	0,15 (0,02-1,61)	9	65	89 / *737*
		2,5	0,11 (0,01-1,18)			
20.04.00 Day 52	FSF (8,0)	10	0,33 (0,04-3,45)	22	44	75 / *506*
		2,5	0,22 (0,04-2,35)			
	KN (6,9)	10	0,20 (0,04-2,43)	16	57	104 / *723*
		2,5	0,09 (0,02-0,97)			
01.06.00 Day 94	FSF (11,0)	10	0,34 (0,03-2,46)	26	49	86 / *422*
		1,0	0,13 (0,03-1,22)			
	KN (10,2)	10	0,14 (0,02-2,03)	24	64	97 / *456*
		1,0	0,05 (0,01-0,89)			
14.09.00 Day 21	FSF (4,5)	10	0,17 (0,05-2,69)	25	58	80 / *960*
		1,0	0,08 (0,03-0,70)			
	KN (4,4)	10	0,15 (0,06-1,32)	21	67	78 / *851*
		1,0	0,06 (0,04-0,43)			
03.11.00 Day 81	FSF (9,2)	10	0,52 (0,08-2,93)	21	52	61 / *358*
	KN (8,7)	10	0,18 (0,02-2,52)	15	70	83 / *458*
23.11.00 Day 101	FSF (10,4)	10	0,62 (0,08-7,83)	21	49	61 / *317*
		1,0	0,13 (0,01-1,80)			
	KN (10,2)	10	0,21 (0,03-1,31)	15	61	90 / *424*
		1,0	0,07 (0,02-0,45)			
25.01.01 Day 10	FSF (3,6)	10	0,44 (0,06-3,72)	21	50	35 / *525*
		2,5	0,11 (0,01-1,21)			
	KN (3,0)	10	0,12 (0,00-1,37)	11	69	75 / *1200*
		2,5	0,03 (0,00-0,36)			
14.02.01 Day 30	FSF (5,5)	10	0,57 (0,10-5,04)	23	42	38 / *373*
		2,5	0,20 (0,06-1,11)			
	KN (4,7)	10	0,24 (0,04-2,02)	11	60	82 / *837*
		2,5	0,08 (0,04-0,58)			
09.03.01 Day 53	FSF (7,7)	10	0,46 (0,06-3,98)	23	45	57 / *400*
	KN (6,8)	10	0,16 (0,01-2,06)	16	62	96 / *678*
19.04.01 Day 94	FSF (11,2)	10	0,91 (0,14-8,65)	21	38	57 / *275*
	KN (9,7)	10	0,25 (0,04-4,10)	10	63	84 / *416*

Livestock unit (LU) = 500 kg liveweight; PM = Particulate matter
FSF = Fully slatted floor, forced ventilation; KN = Kennel housing, natural ventilation

Heber et al. (1988), in studies performed on 11 finishing buildings, with both natural and fan ventilation (where the mean total mass concentration was 8,1 mg/m³), found that the concentration of the total mass and the number of particles are among others negatively correlated to the internal relative humidity, oscillating between 27 % and 100 %. The relative humidity directly affects the generation and retention of aerial dust.

It is assumed that at a relative humidity of 70 % the particles will contain bound and condensed water, which may cause the particles to aggregate together. Wet surfaces and floors again take effect, when the settled dust is bound on the floor and may not become airborne after animal activity (Takai et al., 1998).

However additionally has to be considered, that a swelling effect of high air humidities on hygroscopic particles and there from possible changes in size distribution and behavior of the particles (Seedorf et al., 1999) might affect the measuring accuracy of e.g. optical dust measuring devices.

Trials between kennel housing and partly and fully slatted floor systems for fatteners by Mayer a. Hauser (1999) showed mainly lower dust concentrations for the kennel housing. Additionally depending on the seasonal differences in the ventilation rate in winter increasing dust concentrations for the insulated housing systems were observed.

According to a literature survey by Gustafsson (1999a) there is little consensus among investigations about the influence of the airflow rate or ventilation system on dust concentration. Wang et al. (2000) found that increasing ventilation rates within a certain range resulted in a decrease of the overall mean dust concentration. Heber et al (1988) showed an influence of temperature difference between the stable and outside air (indirect indicator of ventilation rate).

In the frame of in total 256 field studies at different pig buildings in England, the Netherlands, Denmark and Germany presented by Takai et al. (1998), a mean inhalable dust concentration (c.f. section 3.2) of 2,19 (1,87-2,76) mg/m³ and mean respirable dust concentration (c.f. section 3.2) of 0,23 (0,18-0,26) mg/m³ was determined. For pigs kept on slats higher dust concentrations than for pigs kept in litter systems are reported. A seasonal factor was found with higher dust concentrations in winter than in summer for mechanically ventilated buildings, because of normally lower ventilation rates in winter resp. higher ventilation rates in summer. Also the interaction between country and housing type showed significant effects on dust concentration. It is assumed, that environmental factors, such as ventilation parameters, feeding practices, bedding materials, dung and slurry handling can affect dust concentrations.

Finally it is important to understand, that a deduction or calculation of PM emissions from the two housing systems based on the indoor dust concentrations is not permissible. There is evidence that the dust mass concentration and also particle number distribution changes on the way from indoors to the exhaust air opening. I is to be expected because of modifications of the particle properties on grounds of physical effects during transport like among others an additional particle release, settling of dust, or smashing of particles. In the same way alterations like swelling of hydroscopic particles and changes of the chemical composition have been observed (Schneider et al., 2001; Martinec et al., 2000).

For the calculation of PM emissions only dust concentrations measured at the exhaust air opening itself should be taken into account. In this case also an isokinetic air sampling is very important and should be aimed for to ensure accurate measurements. A simultaneous and continuous recording of the respective airflow rate should be striven for, too.

5 Conclusions

The diurnal course and mean indoor dust mass concentration of a housing system for fattening pigs with fully slatted floor, forced ventilation (FSF)and a kennel housing system with natural ventilation (KN) were compared during three to four measuring days of four subsequent fattening periods in the time between autumn 1999 and spring 2001. The main influence on the diurnal course was the animal activity, which itself mainly was determined by the feeding strategy. The mean PM 10, PM 2,5 and PM 1,0 dust mass concentration as well as median and maximum values were in all cases significantly lower for the KN housing system. The higher air humidity, higher share of damp surfaces and on average higher airflow rates are considered to be the main reasons for the over all clear lower indoor dust mass concentration in the KN housing system. Within each housing system a significant negative correlation factor between the mean PM 10 dust concentration and air flow rate per livestock unit could be determined. Because of very likely alterations of the particle characteristics, mass and number concentration in the exhaust air compared to the indoor air, a calculation of PM emissions from the described results of the indoor dust concentrations is not permissible. A calculation of PM emission requires an isokinetic dust sampling at the exhaust air opening with a desirable continuous and simultaneous recording of the air flow rate.

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Mass balances and dust reduction methods in pig houses

Gösta Gustafsson [1]

Abstract

The mass balance of dust as well as the efficiency of different dust reducing measures have been investigated and analysed.

Investigations have shown that the generation of dust is influenced by the number and the weight of the pigs. Settling of dust is a more important mechanism in the mass balance of dust than ventilation rate. A major part of the generated dust settles on different surfaces inside the buildings. The settling rate of dust is affected by the concentration of dust in the air. The settled amount of dust also stands in relation to the floor area of a stable. An increased ventilation rate has a limited effect on the concentration of total dust due to the importance of the settling of the dust. However, it has been observed that the type of ventilation technique may influence concentration of respirable particles. Also the type of housing system influence the generation of dust. One factor which has a strong influence on the concentration of dust is the activity in the buildings.

Dust reducing measures as electrostatic air cleaning of the air and removal of dust with vacuum cleaners have limited influence on dust concentration. Automatic spraying of small droplets of water has reduced the dust concentration with two types of spraying nozzles. For another type of nozzle the generation of dust has increased due to an ultra sound which created an increased activity of the pigs. Spraying with a mixture of rape seed oil has also been effective with manual spraying as well as with an automatic spraying system. The oil seems to have an effect on the generation of dust from the skin but also to function as a dust binding agent for settled dust.

Keywords: Dust, swine, ventilation

1 Introduction

The presence of dust in pig houses may create working environmental (Donham 1987; Tielen et al 1995; Takai & Iversen 1990; Larson et al 1993; Malmberg et al 1993) problems as well as depressed health status of the animals (Donham 1991; Robertson et al 1990; Robertson 1993; Hamilton et al 1993). Measures to reduce the contamination of the air in swine confinement houses are therefore urgent. The purpose of these investigations has therefore been to analyse the mass balance of dust but also the effectiveness of different dust reducing measures.

The major part of swine house dust is organic. Originally, the dust was considered to origin from feedstuffs. However, investigations (Hartung 1992) have indicated that there are also other components of the dust as particles from skin, hair and faeces. Investigations (Angst 1984; Hartung 1992) have shown that the composition of settled dust and feedstuffs in pig houses differ considerably regarding crude protein and crude ashes.

The major part of the number of dust particles are respirable (Nilsson 1982). However, it should be observed that the major part of the weight of the dust is not respirable. Donham (1986) reported that 7 % of the total weight of the dust was respirable.

A considerable proportion of the dust seems to originate from the pigs themselves. Nilsson (1982) found that the type of feed (dry or wet) had limited influence on the daily averages of total dust concentrations in growing-finishing pig houses. However, both in cases with wet and dry feed the dust concentrations increased during the feeding time due to an increased activity.

Several investigations (Nilsson 1982; Gustafsson 1994; Pedersen 1993; van't Klooster et al 1993) have proved that the activity in swine houses has a strong influence on the concentration of dust in the air. The concentration normally increases during periods when the activity is high, such as during feeding, weighing of the pigs, etc. The influence of feeding technique on the activity of the pigs may have an indirect effect on the dust concentration (Robertson 1992). Pedersen (1993) has shown that the number of dust particles in the air varies with the same pattern as the signal from an activity sensor.

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There is little consensus among investigations about the influence of ventilation on dust concentration. However investigations (Bundy & Hazen 1975; Bundy 1984) about the influence of ventilation rate on the number of dust particles show a decrease in number of dust particles at increasing air flow rate. The influence of ventilation rate on total mass concentration of dust in the air has been less pronounced (Nilsson 1982; Gustafsson 1994). However, influence of temperature difference between the barn and outside air (indirect indicator of ventilation rate) have been found (Heber et al 1988). Investigations have also indicated influence of different ventilation techniques on dust concentrations (van't Klooster et al 1993).

2 Theory

The mass balance of generated dust has been described by Nilsson and Gustafsson (1987) as:

$$V\frac{dC_{av}}{dt} = p - q(C_o - C_i) - SA - G \qquad (1)$$

where: V is the building volume in m^3 ; C_{av} is the average dust concentration; C_o and C_i are the total dust concentrations in outlets and inlets in mg/m^3 ; t is time in h; p is the production of dust in mg/h; q is the ventilation rate in m^3/h; S is the settling rate of dust in mg/ m^2 h; A is the area of the floor in m^2; and G is the amount of dust removed by air cleaning devices in mg/h.

The settling of dust may be described by:

$$S = v\, C_{av} \qquad (2)$$

where v is a value depending on the properties of the dust in m/h.

If stationary conditions are maintained, is it possible to determine the generation of dust from:

$$p = q\,(C_o - C_i) + S\,A + G \qquad (3)$$

The fraction removed by air cleaning devices is described by:

$$G = \theta\; q\; \eta\; C_{av} \qquad (4)$$

where θ is the relation between the air flow rate of an air cleaner and the ventilation rate of a barn and η is the air cleaning efficiency of an air cleaning device. The concentration in the inlet of the air cleaner is assumed to be the average dust concentration in the air C_{av}.

3 Material and Methods

3.1 Buildings and equipments

The investigations have been carried out in three piggeries for growing-finishing pigs at the research station Alnarp Södergård.

The influence of the following factors in the building environment has been investigated, namely: number and weight of animals; activity; settling of dust; ventilation rate; ventilation technique; and animal housing system.

The following methods to reduce the generation and concentration of dust have also been investigated, namely: electrostatic air cleaning; dust removal by vacuum cleaning; humidification of the air with different spraying nozzles; and oil treatment.

3.2 Measurements

The efficiency of different treatments have been analysed by: gravimetically measurements of the amount of total dust in mg/m^3 with 37 mm diameter dust filters (Millipore) at a flow rate of 1.9 l/min located in the middle of the barn at 1.5 m height but also in the exhaust air; gravimetically measurements of the amount of respirable dust (mg/m^3) with dust filters (Millipore) after separation of particles larger than 5 μm with a cyclon (SKC cyclon) at the same locations as for total dust; counting the number of particles of different sizes with an optical particle counter (Rion) which counted the number of particles of size larger than 0.3, 0.5, 1.0, 2.0, and 5.0 μm; weighing settled dust on five 0.230 m^2 settling plates located at a height of 2.0 m with the collected amount of dust measured by weighing the plates on a balance; measuring the ventilation rate with a hot wire anemometer (Alnor) in the exhaust air ducts.

Each measurement was carried out over a period of 3-4 days in order to collect enough dust on the settling plates. Different treatments have been compared to reference values measured before and after the treatments.

3.3 Analyses

Different measures to reduce the generation and concentration of dust have been analysed by using the following properties in the mass balance Eqn (1): averages of total and respirable dust concentrations C_{tot} and C_{resp} measured in the middle of the barn and in the exhaust air; average of settling rate of dust on settling plates S; generation of dust p as defined by Eqn (3); relation between settled amount of dust and

dust concentration S/C_{tot}; and fraction of respirable dust C_{resp}/C_{tot}.

Measurements of the number of particles have mainly been used to get a picture of the particle size distribution and influence of activity and ventilation rate.

4 Results and Discussion

The influence of number of pigs on production of dust was investigated by changing the number of pigs when their average body weight was in the range of 86 – 98 kg. The measurements showed that the generation of dust is proportional to the number of animals (figure 1).

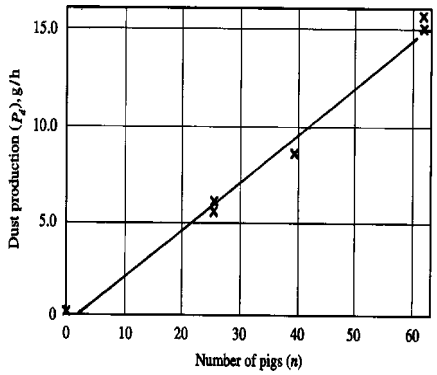

Figure 1:
Relation determined between the production of dust and the number of pigs.

The influence of pig weight on dust production was also investigated during 14 production batches with growing finishing pigs. The production of dust increased with the body weight in all batches. The influence of body weight on dust production has therefore been analysed by linear regression which is presented in figure 2.

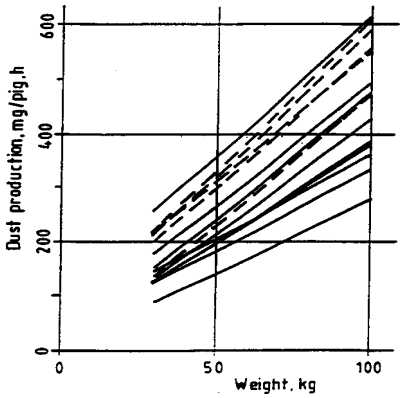

Figure 2:
Relations determined between the production of total dust and average pig weights during 14 production batches with growing finishing pigs.

The settling rate of dust varies to a large extent between different locations inside the buildings. However, it has also been found that the variations in the settling rate follow the same pattern over the entire production period. This fact indicates that the air flow patterns inside the buildings could have an influence on the dust conditions.

An example of relation between settling rate S and the total dust concentration C_{tot} is presented in figure 3. Determinations indicate that the settling rate of the dust is influenced by the concentration of dust in the air.

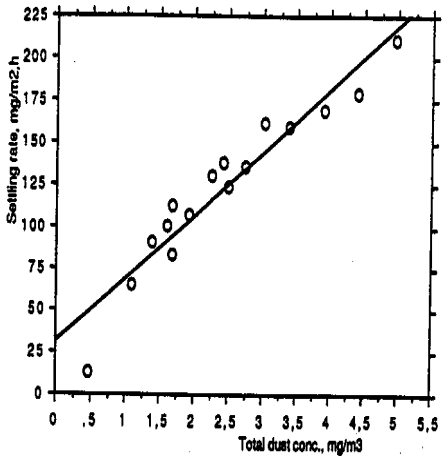

Figure 3:
Relations determined by linear regression between the settling rate and the concentration of total dust during a production batch with growing- finishing pigs.

Increased ventilation rate is often recommended as a method to reduce the concentration of air pollutions in buildings. Unfortunately, the ventilation

rate has a limited diluting effect on dust at those ventilation rates recommended for insulated swine houses in temperate areas. The reason is that the settling of dust on different surfaces is a more important mechanism to remove dust particles from the air than the ventilation rate in pig houses. The major part of the dust settles on different surfaces inside the buildings. Figure 4 shows an example of the limited effect on total dust concentration at different ventilation rates. The dilution of the dust by increased ventilation will increase the heating requirement in temperate regions.

The fraction of the generated dust which is exhausted by the ventilation air is presented in figure 5. The fraction of the dust which is exhausted is limited at those ventilation rates which occur in swine confinement houses in temperate areas. The low fraction of exhausted dust shows that the settling of dust is more important than ventilation rate in the mass balance of dust.

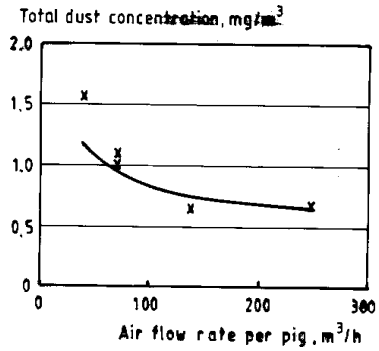

Figure 4:
Example of influence of ventilation rate on total dust concentration.

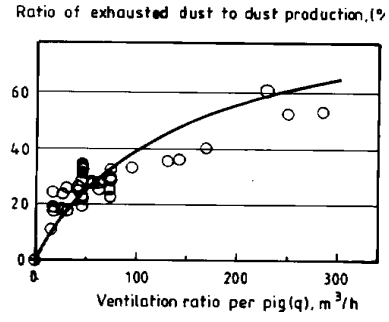

Figure 5:
Ratio of dust extracted by the ventilation system to dust production.

The influence of ventilation rate on the number of dust particles of different sizes when air has been supplied with a high speed recirculating air inlet is presented in figure 6. The ventilation has had a diluting effect mainly on particles larger than 1.0 μm. The ventilation rate had no effect for particles smaller than 1.0 μm for this particular ventilation system.

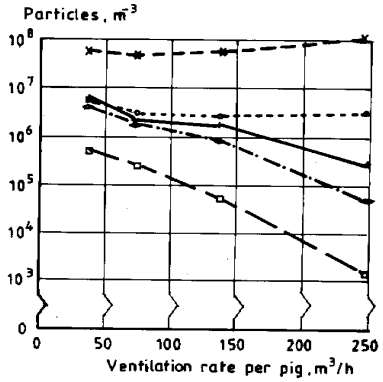

Figure 6:
Influence of ventilation rate on the number of dust particles of different sizes; x ___ ___ x, 0.3-0.5 μm; o _ _ _ _ o, 0.5 - 1.0 μm; +_____+, 1.0- 2.0 μm; ♦ __ . __ ♦, 2.0- 5.0 μm; ____ ____, > 5.0 μm

Two very different ventilation principles have been compared, namely: high speed recirculating air inlets in combination with an exhaust fan located at roof level (high exhaustion); and a porous ceiling as the air inlet in combination with manure gas ventilation (low exhaustion).

The recirculating air inlets create considerable air mixing and air movements in the stable while the air movements from the porous ceiling are extremely small.

Experimental data for dust concentration and production, settling rate, ratio of respirable dust and fraction of exhausted dust are presented in Table 1. Significant differences ocurred regarding respirable dust concentration C_{resp} and S/C_{resp}. These results indicate that the ventilation technique (mainly air velocities and air movements) may have an influence on small particles.

Two different housing systems were compared namely: climate controlled confinement in an insulated piggery; and cold confinement in an uninsulated piggery with straw bedding and natural ventilation.

In all investigated batches except one, significant differences occured between the different piggeries, see Table 2. The presence of dust was much lower in the uninsulated stable with strawbedding. The reasons to the large differences between the different systems are difficult to explain. Possible explanations may be more moisture in the cold environment with straw bedding, and very high ventilation rates during spring, summer and autumm in the uninsulated stable.

Table 1:
Total and respirable dust concentration C_{tot} and C_{resp}, dust production, ratio between settling rate *S* and total and respirable concentrations, ratio of respirable to total dust concentration and fraction of exhausted dust at two different ventilation techniques

Parameter	High speed air inlet + high exhaustion			Breathing ceiling +low exhaustion			Difference	
	x	s.d.	n	x	s.d.	n	%	
Total dust conc., mg/m^3	1.29	0.57	10	1.14	0.32	7	+13	NS
Resp. dust conc., mg/m^3	0.26	0.095	6	0.15	0.061	4	+77	*
Dust prod. per pig, mg/h	253	104	11	322	116	6	-21	NS
S/C_{tot}, m/h	86	41	11	69	12	7	+24	NS
S/C_{resp}, m/h	392	86	6	535	80	4	-27	*
Ratio of resp. dust, %	18.8	3.5	6	14.2	4.8	4	+32	NS
Exhausted dust, %	25.9	7.3	9	21.0	6.0	5	+23	NS

x, average; s.d., standard deviation; n, number of batches; N.S., non-significant difference;
*, significant difference 0.05 >p >0.01

Table 2:
Total and respirable dust concentration C_{tot} and C_{resp} and settling rate of dust *S* at five comparative production batches with growing-finishing pigs in two different housing systems

Property	Trial	Insulated and climate controlled			Cold confinement with strawbedding and natural ventilation			
		x	s.d.	n	x	s.d.	n	Δ
Total dust conc. mg/m^3	1	1.26	0.57	10	0.19	0.06	15	-75 ***
	2	1.91	0.82	17	0.91	0.22	11	-52 ***
	3	1.00	0.40	14	0.39	0.1	14	-61 ***
	4	0.787	0.35	17	0.62	0.41	7	-21 NS
	5	1.37	0.59	23	0.45	0.15	23	-67 ***
Resp. dust conc. mg/ m^3	2	0.30	0.23	5	0.096	0.087	10	-68 *
	3	0.09	0.05	11	0.034	0.036	13	-62 **
	4	0.14	0.07	16	0.215	0.146	6	+53 NS
	5	0.15	0.063	23	0.059	0.015	7	-61 ***
Settling of dust mg/m^2, h	1	67	22	21	30	20	20	-55 ***
	2	72	27	17	45	29	9	-38 *
	3	55	24	15	30	8	14	-45 ***
	4	63	25	17	63	25	20	0 NS
	5	71	22	23	52	21	22	-27 ***

x, average; NS, non-significant difference; s.d., standard deviation; n, number of batches;
Δ, difference %; *, significant difference 0.05 >p >0.01; **, significant difference 0.01 >p >0.001; ***, significant difference 0.001 >p

The use of an electrostatic air cleaner had a limited effect on the dust concentration in the air, although it has been proved that the equipment removed a large fraction of the particles in the air which passed through the device. Considering the mass balance of the dust, it is obvious that air cleaning devices need large airflow capacities if the dust concentration in the air is to be affected. The airflow through an air cleaner has the same influence on the dust concentration as an equally large increase in ventilation rate in the building.

The use of a vacuum cleaner designed for industrial purposes, as well as a central vacuum cleaning system, have been investigated. Both devices have been used to clean floor surfaces but also other surfaces such as pipes, etc. at different cleaning intervals. Although most surfaces looked cleaner after the treatments, no significant effect could be measured regarding total and respirable dust concentrations, settling rate or generation of dust.

Three types of spraying nozzles have been investigated in an automatic spraying system namely: high pressure (ultra sound) nozzles; flat fan nozzles; and full cone nozzles.

The nozzles have been operated automatically in short sequences. They were operated twice per hour from 8 a.m. until 6 p.m. and once per hour during the rest of the day.

Spraying water droplets have given different results dependent on the type of nozzles which have been used. The use of ultra sound nozzles which created droplets in the size range between 5 and 10 μm resulted in a significant increase of both total and respirable dust concentrations during nine comparative trials. The reason for the increased dust concentrations was probably the ultra sound (frequency 30 kHz) created by the nozzles. This sound was outwith the human hearing range. However, observations of the pigs clearly showed that the pigs reacted in an abnormal way the first times the nozzles were in operation. The increased dust concentrations may only be explained by an increased activity of the pigs due to the ultra sound.

The use of the flat fan nozzles operated with a pressure of 0.35 MPa gave a reduction in both total and respirable dust concentrations. In these trials, each pen was equipped with four (horizontal spraying direction) flat fan nozzles in combination with a full cone nozzle (orientated downwards).

The use of full cone nozzles operated at 0.3 MPa pressure also reduced both total and respirable dust concentrations. The settling rate and the generation of dust were also affected. The efficiency was improved with increasing length of the spraying periods, see figure 7.

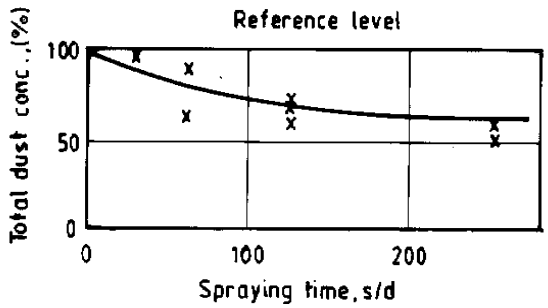

Figure 7:
Relative change in dust concentration in % when different amounts of water were supplied with full cone nozzles

It has earlier been proved by Takai et al. (1993) that the spraying of mixtures of oil and water in pig houses will give a significant reduction in dust concentrations. However, it has not been verified whether the reduction of dust is due to less generation of dust from the pigs skin surfaces or if the oil functions as a dust binding agent on different building surfaces.

In these investigations, 10% rape seed oil in a water solution was used. The mixture has been applied in two different ways namely: manually spraying directly on the pigs with a knapsack sprayer; and automatically with a spraying system with full cone nozzles parallel to the feeding troughs. In the latter case, the oil mixture was applied once per day during the feeding time.

The manual treatment affected all the parameters measured. In order to see if the oil affected the release of dust from the skin, one treatment was carried out outside the building so that no oil should cover any building surfaces. In this treatment, the total dust concentration was reduced to 84% of the reference level. The treatment had a significant reduction on settling rate (63% of the reference level) and generation of dust (72% of the reference level). It can be concluded that the treatment with oil has reduced the generation of dust from the skin to some extent but also that the oil treatment functions as a dust binding agent on surfaces in the building.

An automatic system for spraying of oil has also been investigated. The automatic spraying system consists of two full cone nozzles per pen located parallel to the feeding troughs. The oil mixture was sprayed over the pigs back once per day during the feeding of the pigs. The reduction on total dust concentration at different amounts of oil is presented in figure 8. The treatments resulted in a considerable reduction in total dust concentration. Reduction levels in the range of 75-80% has earlier been reported by Takai et al. (1993) with a high pressure spraying system.

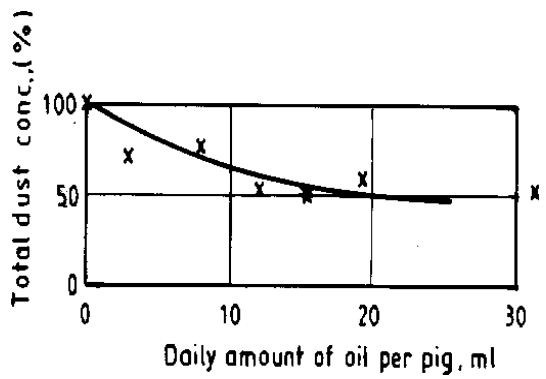

Figure 8:
Relative change in total dust concentration in % when different amounts of oil were supplied with full cone nozzles.

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Properties of airborne dust from pig husbandries and their effects on measurement

Friedhelm Schneider, Thomas Engelhardt, Paul Horst Wieser [1]

Abstract

When measuring dust concentration inside an animal husbandry and the emission of aerial dust into the environment optical particle counters become more and more popular. The Dust Monitor Model 1.108 (GRIMM Inc.) represents a relatively robust, handy and cheap portable instrument, suitable for continuous "on line" measurements of the particle number concentration and size distribution.

This device has been used with measurements in a special experimental swine housing to quantify time series of particle number concentration with high temporal resolution. Maxima in the particle number concentration occur periodically in the covered size range 0,3µm to >20µm (D_o optical latex equivalent diameter). They are significantly correlated with the feeding procedures.

The performance of the instrument was evaluated, taking into account the hardware of the OPC and the Mie-theory of light scattering of small particles. The estimation of the accuracy of the instrument was the major task of this theoretical modeling. The optical properties of dust particles inside a piggery are known insufficiently. Therefore a set of indices of refraction according to transparent, low and high absorbing particle materials has been assumed. The results clearly indicate the substantial influence of the refractive index on the optical determination of the particle size, in particular for strong absorbing particles.

Introduction

The dust concentration inside an animal husbandry and the emission of aerial dust into the environment by ventilation are important items for the validation of animal welfare and environmental hygiene. The selection and use of a dust measuring device depends on the particle number concentration and size distribution, the temporal and spatial fluctuation of these characteristic quantities and last but not least on the financial conditions.

The concentration of dust particles inside animal husbandries varies diurnal and according to the season due to the time dependent activity of particle sources and sinks. This has been shown with different animals and under varying conditions in many publications (e.g. Müller and Wieser, 1987; DeBey et al., 1994; Hinz and Linke, 1998; Pedersen, 1998).

Consequently continuous "on line" measurements are particularly suitable and in most cases necessary for the study of temporally variable particle sources and sinks. These kind of measurements can be carried out with sufficient temporal resolution by optical particle counters (OPC). The Dust Monitor Mod. 1.108 (GRIMM Inc.) is a relatively robust, handy cheap and meanwhile popular device. (weight 2.5 kg, dimensions 24 x 12 x 6 cm, cost approx. 10.000 €, http://www.grimm-aerosol.de/)

The purpose of this work was to evaluate the practicability of the Dust Monitor Mod. 1.108 (GRIMM Inc.) for automatic long-term particle measurements inside animal husbandries and in addition to estimate the influence of optical particle properties on the accuracy of the results received.

Material and methods

The measuring was carried out at a special experimental swine housing located at the research facility 402 “Unterer Lindenhof" of the University of Hohenheim. The swine housing was designed by the Department of Agricultural Engineering, Process Engineering in Animal Production and Farm Structures (Prof. Dr. T. Jungbluth), University of Hohenheim. The fully slatted floor system (FSF) is divided in 6 pens with 9 animals each. The pen-size without trough area averages 3.30m x 2.20 m (0.8m²

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net area per animal). The resting area is uncovered on fully slatted concrete floor. Each pen has a single transverse trough with one occupation technique. The liquid feeding, with a number of 16 fixed feeding times from 6:00 to 22:00 is controlled by a feed level sensor in the trough. The feed mixture consists of bruised barley, wheat, triticale, oats, soy extract, soy oil, mineral additives and calcite in different mixing ratios, depending on the age and weight of the pigs. At 15:00 and 22:00, a so-called turbo cleaning procedure blows compressed moist air through the feeding pipes to avoid sticky contaminations. The FSF-system has a forced ventilation, central pore canals over each pen row for inlet air with a total surface area of 24m² and a underfloor extraction below the entire feed passage. The air leaves the compartment through one exhaust air chimney, 7m high and 0.6m in diameter. The slurry container for liquid demanuring, under each side of the feeding passage has a storage capacity for one fattening period. More detailed information about the complete design and the installed measurement technique was published e.g. by Gallmann et al. (2000a, 2000b).

The Dust Monitor Model 1.108 (GRIMM Inc.) measures particles within a size range of 0.3µm < D_o < 20µm in diameter (D_o: optical latex equivalent diameter), and gives information about the size distribution by classifying the signals into 15 size channels. The lower class limits of the size channels are 0.3, 0.4, 0.5, 0.65, 0.8, 1, 1.6, 2, 3, 4, 5, 7.5, 10, 15, 20µm, respectively. The controlled flow rate was 1.2 liter/minute. The OPC was equipped with a sensor for humidity, temperature and air velocity. The device was mounted inside a specially designed box with power supply, air inlet 1.2m above the floor and air outlet tube. The device was placed in the service alley, near the center of the building. The measuring interval for all 15 size channels and the sensor data was 6 seconds. Mean values of 10 measurements were stored every minute for maximum 7 days continuously.

OPCs use light scattered by the particles for particle identification and size classification. They measure partial scattering cross sections from single particles appearing in the view volume of the light detection system (e.g. Willeke and Liu 1975). The partial scattering cross section depends on spectral properties of the light source, measured scattering angle, particle size, particle shape and on the particles index of refraction. The particle size is indicated as a diameter of a spherical particle with the same partial scattering cross section as the standard particle used for calibration. Polystyrene latex spheres are a common particle standard. When polystyrene latex spheres are used for calibration, the OPC determines optical latex equivalent diameters D_o.

Based on Mie-theory and the hardware of the Dust Monitor Mod. 1.108 (GRIMM Inc.) the OPC response was modeled to estimate the accuracy of particle sizing. Refractive indices (m_i), particle size (D_o), scattering angle 90° (46,45< Θ < 133,35) and wavelength of the laser diode (λ = 780 nm) are the input data in the Mie-model. The Mie-calculations (Bohren and Huffman 1998) were carried out for homogeneous spheres by use of the software "MieCalc" (Dr. Bernhard Michel Scientific Consulting, Feucht, Germany). Taking into account, that the Dust Monitor Mod. 1.108 (GRIMM Inc.) derives information about the particle mass-distribution from the measured particle size distribution, using an estimated density of the particle material, the investigations were restricted to study the influence of the refraction index on the accuracy of particle sizing. Detailed information about the optical properties of stable dust depending on the chemical nature and the degree of internal and external mixing of the main particle components are missing. Therefore a set of five spherical, homogeneous particle types characterized by defined indices of refraction has been assumed. As shown in tab. 1, the indices of refraction stand for transparent, low and high absorbing particle materials.

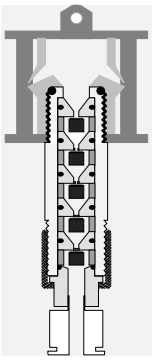
Table 1:
Particle types and their refractive indices

Particle type	Refraction index m_i	
1	1,59 – 0i	Polystyrene-latex[1]
2	1,52 – 0,004i	Mineral dust
3	1,49 – 0,009i	Sulfate
4	1,56 – 0,086i	Urban aerosol
5	1,74 – 0,43i	Soot

[1] Duke Scientific Corporation, Palo Alto, California

In addition, microscopic pictures of coarse dust particles from inside the piggery have been taken, to evaluate particle shapes. Single jet five-stage mini-cascade impactors (MKI-S5) were used to collect particle samples, suitable for single particle micro analytic techniques.

Table 2:
Dimensional sketch of the MKI-S5 with an PM 10 sampling head, 50% cut off diameter D_{50} (aerodynamic latex equivalent diameter) of the five impactor stages, nozzle diameter and average nozzle exit velocity.



Stage	Nozzle diameter W / µm	50% cut-off-diameter D_{50} / µm	nozzle exit velocity v / ms^{-1}
1	1300	3,5	9
2	800	1,2	23
3	600	0,65	41
4	400	0,35	93
5	300	0,18	165

These impactors have been designed at the Institute of Physics, University of Hohenheim (Wieser and Wurster, 1986). The 50% cut off diameter D_{50} (aerodynamic latex equivalent diameter) of the five impactor stages, the nozzle diameter and the average nozzle exit velocity are given in tab 2. The dimensional sketch in tab. 2 shows the MKI-S5 with an PM 10 sampling head. In each impactor stage particles were deposited on a Pioloform® coated hexagonal mesh nickel grid (Inc. PLANO). The thickness of the transparent Pioloform® coating is approx. 20nm. The grids, used for transmission electron microscopy, have a diameter of 3.05 mm and are available with different number of meshes (mesh = bars per inch). They are magnetically fixed on an impaction-plate. For the single particle analysis no further preparation was necessary.

Results and discussion

As illustrated in figure 1 the 48 hours' time series of the particle number concentration inside the piggery shows typical periodical fluctuations. The periodically appearing maxima are most significantly linked to the feeding intervals, which are characterized by high animal activity again.

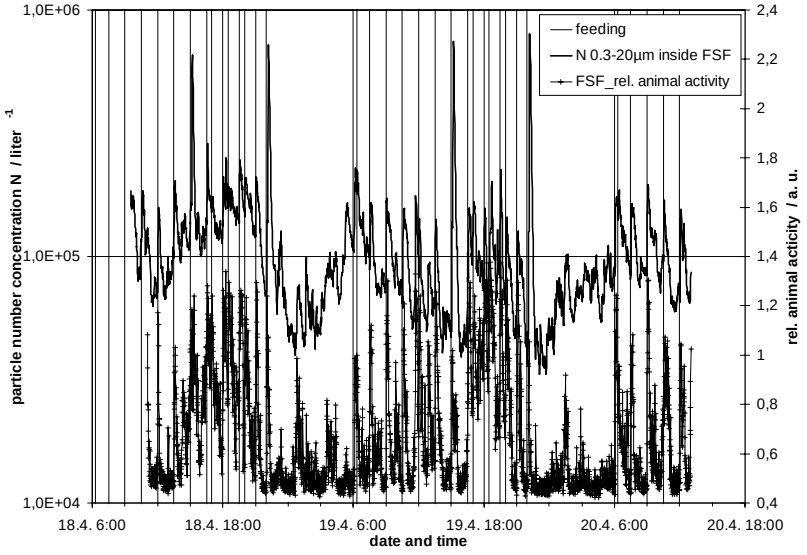


Figure 1:
2 days' time series of particle number concentration inside the FSF system in the size range 0.3µm to 20µm in diameter (optical latex equivalent diameter) in comparison with animal activity. The vertical lines indicate the feeding times.

These effects can be seen in both the submicron and coarse particle size ranges. In order to demonstrate this results figure 2. shows exemplarily for the size channel 1 (0.3µm to 0.4µm in diameter) and size channel 14 (15µm to 20µm in diameter) four hours' time series of the same data set. Comparing the left and right part of figure 2, the different order of magnitude of the particle number concentration should be noticed. A comparison of the two particle number concentrations indicates a strong dependence of the particle number concentration on particle size. Surprisingly, the effect of the turbo cleaning procedure at 22:00 o' clock is distinctively high in the size channel 1, however not observable in the size channel 14. Consequently, the cleaning procedure of the feeding system is a temporary strong source of small particles.

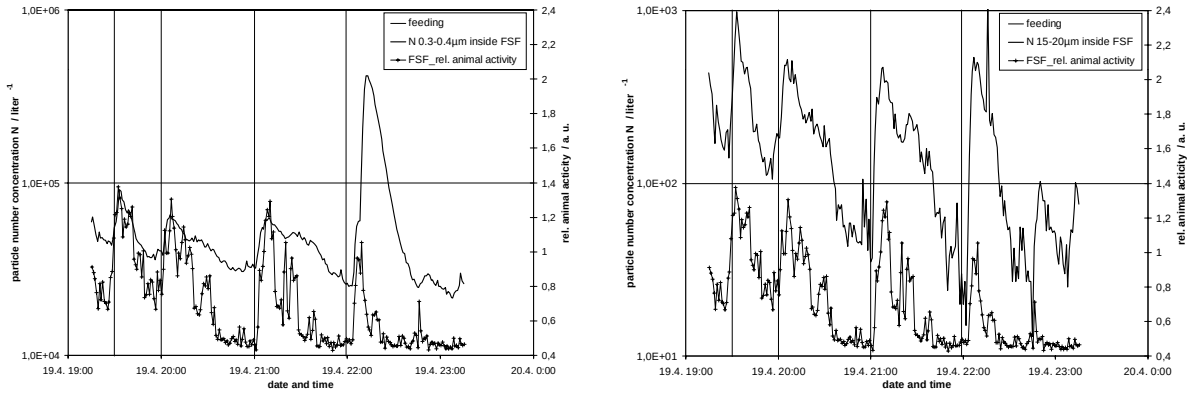


Figure 2:
4 hours time series of particle number concentration inside the FSF system in the size channel 1 (0.3µm to 0.4µm in diameter, left part) and size channel 14 (15µm to 20µm in diameter, right part) in comparison with animal activity. The vertical lines indicate the feeding times.

The decay of the concentration peaks is affected by stirred particle settling and by ventilation. A reduction of the dust emission in this case should be possible by changing the feeding technique.

Figure 3 shows the calculated relative scattering, depending on particle size for the five different indices of refraction. The vertical dotted lines represent the lower limits of the 15 size channels of the Dust Monitor Model 1.108 (GRIMM Inc.), based on the manufacturers' instructions. The spotted horizontal lines represent the size channel threshold of the relative scattering intensity, based on the calculated relative scattering of the polystyrene latex standard spheres.

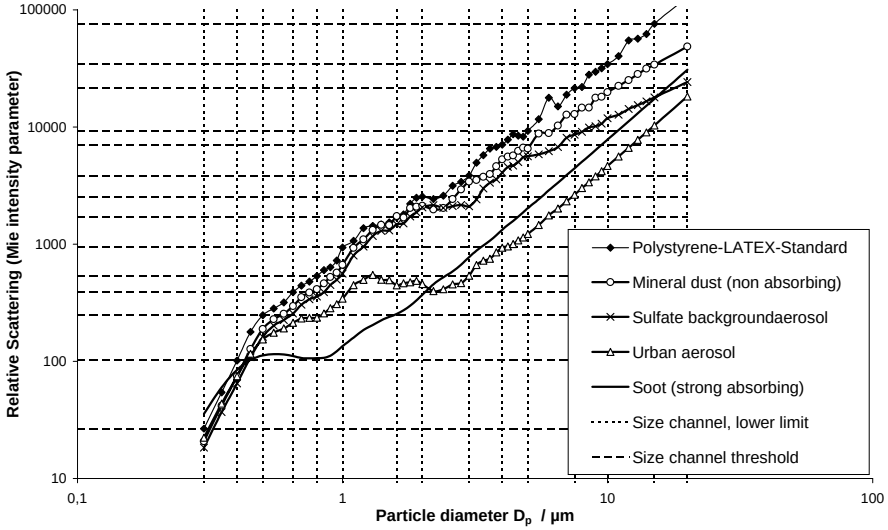


Figure 3:
Dependence of relative scattering (Mie-theory) and particle size for five different particle types.

The relative scattering as a function of the particle diameter depends on the refractive index. These results clearly indicate the substantial influence of the refractive index on the optical determination of the particle size, in particular for strong absorbing spheres.

The size channels are chosen by the manufacture that way, that the particle size classification seems to be unambiguous for polystyrene latex. As shown in tab. 3 the real geometric particle diameter of particle types with different indices of refraction is shifted in comparison to the latex equivalent diameter. This effect increases both with particle size and absorption. In this study the model calculations are limited to cases with real geometric particle size diameters lower than 20µm.

Table 3:
Size ranges (µm) of the five particle types (given in tab. 1) classified in the 15 size channels of the dust monitor mod. 1.108 (GRIMM, Inc.), depending on particle typs' index of refraction.

	Particle typs' size ranges / µm				
Size channel	**1**	**2**	**3**	**4**	**5**
1	0,3 - 0,4	0,3 - 0,43	0,3 - 0,44	0,3 - 0,43	0,25 - 0,44
2	0,4 - 0,5	0,43 - 0,58	0,44 - 0,62	0,43 - 0,83	0,44 - 1,5
3	0,5 - 0,65	0,58 - 0,75	0,62 - 0,86	0,83 - 1,1	1,50 - 2
4	0,65 - 0,8	0,75 - 0,92	0,86 - 0,99	1,1 - 1,05	2,0 - 2,4
5	0,8 - 1	0,92 - 1,1	0,99 - 1,2	1,05 - 4	2,4 - 3,3
6	1 - 1,6	1,1 - 1,7	1,2 - 1,8	4 - 5,8	3,3 - 4,5
7	1,6 - 2	1,7 - 2,6	1,8 - 3,25	5,8 - 7,3	4,5 - 5,6
8	2 - 3	2,6 - 3,5	3,25 - 3,9	7,3 - 9	5,6 - 6,9
9	3 - 4	3,5 - 5,1	3,9 - 6,5	9,0 - 12,4	6,9 - 9,5
10	4 - 5	5,1 - 6	6,5 - 8,2	12,4 - 14,5	9,5 - 11,1
11	5 - 7,5	6 - 10,4	8,2 - 17,5	14,5 - >20	11,1 - 16,5
12	7,5 - 10	10,4 - 15,1	17,5 - >20	>20	16,5 - >20
13	10 - 15	15,1 - >20	>20	>20	>20
14	15 - 20	>20	>20	>20	>20
15	>20	>20	>20	>20	>20

Aerial dust inside this piggery is mainly formed by feed constituents like bruised grain, mineral additives and other biogenic matter (Schneider et al., 2001) Although such materials don not show absorption coefficients comparable to urban aerosol or soot, a significant inaccuracy in the particle size classification of coarse particles is expected when using the Dust Monitor Model 1.108 (GRIMM Inc.).

Microscopic images of coarse dust particles from inside the piggery (figure 4) show, that in the minority of cases the particles match with spherical shapes.

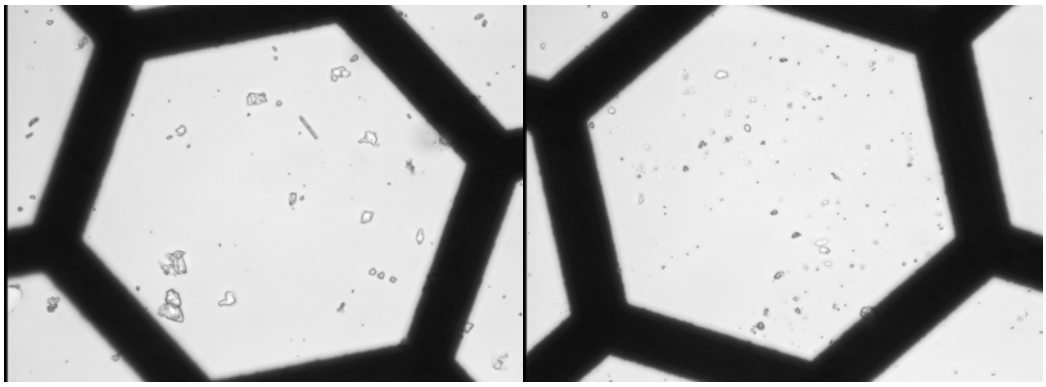


Figure 4:
High resolution light microscopy scan (X 400) of particles sampled inside of the FSF piggery. Left picture shows particles deposited on impactor stage 1 (D_{ae} > 3,5µm), the right picture shows particles deposited on impactor stage 2 (1,2 < D_{ae} < 3,5µm.).

The assumption of spherical particles seems to be a very rough estimation. The difference of geometrical size of dust particles deposited on the same impactor stage indicate different densities of the particle materials. Also the loss of volatile liquid coatings shows the same effect. Consequently experimental data about the real particle shape and inhomogeneous chemical composition should be integrated in further work.

Conclusion

The Dust Monitor Model 1.108 (Grimm Inc.) is calibrated with transparent standard spheres (polystyrene-latex). Consequently the real geometric particle diameter differs from the measured optical equivalent diameter. This inaccuracy depends on particle size and index of refraction. Substantial measuring errors can occur with light absorbing coarse particles.

Specific investigations of optical properties of dust particles are necessary to prevent or evaluate these measuring errors. The optical properties depend e.g. on the chemical nature and the degree of internal or external mixing of the components forming the air particulate matter of representative particle collections. Investigations of dust particles sampled in an experimental piggery are on the way by using laser microprobe mass analysis (LAMMA) and analytical electron- or light microscopy.

The deviation of the dust particles from the spherical shape raises an additional question.

Acknowledgement

We acknowledge Wolfgang Bea from the Institute of Agricultural Engineering of the University of Hohenheim for the assignment for beneficial use of data, especially animal activity.

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This publication is dedicated to Prof. Dr. sc. agr. Dr. h.c. Jürgen Zeddies, University of Hohenheim, Institute for Farm Management, Section Planning and Organisation of Agricultural Production, Speaker of the Graduiertenkolleg 768, on the occasion of his 60th birthday.

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Concentrations of airborne dust in different farrowing systems

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Abstract

Total suspended particulate matter (TSP) was measured in four farrowing compartments, each housing 6 pens for sows kept individually on perforated floors. The only difference between the pens in the 4 compartments was the degree of freedom of movement for the sows: compartment 1, farrowing crates; compartment 2, farrowing crates, opened after the castration of the piglets; compartments 3 and 4, locomotion pens. The experiments were carried out in the context of a research project dealing with animal friendly systems for the housing of sows.

The data of 40 measuring periods were analysed. 11, 9, 10 and 10 of these measurements were carried out in the four compartments mentioned above. The periods consisted of 69 to 744 single measurements after the first parturition. The data analysis was restricted to 672 single measurements per measuring period. The frequency of the measurements was 0.5 Hz. The data analysis was carried out with the rolling average of the dust mass stored with a frequency of 0.56 mHz.

Average concentration of airborne dust in the 4 compartments amounted to 301, 324, 268 and 349 µg m^{-3} respectively (n = 40). The corresponding standard deviations amounted to 86, 86, 55 and 314 µg m^{-3} respectively. Neither the compartment, i.e. the possibility of the sow to move around, nor the number of weaned piglets or the supply of straw to allow nest building behaviour did affect the concentration of airborne dust significantly.

Introduction

According to the report of an expert working group established by the Scientific Veterinary Committee of the EU sows are usually kept in separate units from a few days before expected farrowing until weaning. The housing systems may be divided into two main groups: sows kept in crates or loose sows. In intensive pig production, crate systems dominate (von Borell et al., 1997).

The authors pointed out, that the farrowing crates have both advantages and disadvantages from a welfare point of view. The advantages are: reduced piglet mortality caused by crushing, easier management including better surveillance of the parturition, easier health control and, in case of disease, easier treatment of the periparturient sow. According to von Borell et al. (1997) the disadvantages are: reduced possibilities for the sow to perform nest building behaviour, signs of increased stress before parturition, some studies indicate a decreased risk for MMA in the periparturient sow when kept loose. Furthermore it is disadvantageous that any contact between sow and piglets is made more difficult and that the sow's possibility to move is restricted to a minimum for a period of several weeks.

The expert working group mentioned above concluded that further development of farrowing systems in which the sow can be kept loose and carry out normal nest building without the system compromising piglet survival, should be strongly encouraged.

Taking these reflections as a basis, some alternatives to the common farrowing crate have been installed when modernising the piggery of one experimental farm of Göttingen University. The comparison of these housing systems started at the beginning of 1999, taking parameters of animal welfare, animal health, climate inside the stable, economy and productivity into account. A comparison concerning the working time has been completed already (Snell et al., 2001).

The investigations presented here deal with the question whether allowing the sows to move around in their pen and offering straw to allow nest building behaviour affects the concentration of dust in the compartment and whether further influences on the amount of airborne dust in farrowing houses exist.

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Materials and Methods

Animal Husbandry

The investigations took place within the scope of a scientific project named 'Evaluation of different farrowing pens considering especially animal-welfare and economy'. This project was sponsored by the *Deutsche Forschungsgemeinschaft*, DFG.

The experiments were carried out in the piggery of the experimental farm Relliehausen. Four compartments with 6 farrowing pens each were used. The compartments only differed as far as the construction of the farrowing pens was concerned.

A compartment passage was situated in the middle of each compartment, three farrowing pens (2000 x 2490 mm) facing it at both sides. Standard parts (Laake, Langen, D) were used for construction. The walls of those pens in which the sow could move around freely were heightened from 600 to 1100 mm. Additionally, restraining stirrups were installed at the walls.

The housing systems compared can be characterised as follows:

Compartment 1: Farrowing crate, arranged diagonally, fixing the sow until weaning.
Compartment 2: As compartment 1, but the crate was opened after castration (ca. 10^{th} day), one part forming a farrowing rail next to the wall, the other protecting the nest.
Compartment 3: As compartment 2, but half of the crate was removed, the other part protected the nest; turning this one towards the wall made fixation of the sow possible.
Compartment 4: As compartment 3, but no fixation of the sow was possible (figure 1).

As the device to fix the sow was not used in compartment 3, the housing conditions in the compartments 3 and 4 were identical.

The ventilating system (Fancom, NL) was controlled separately for each compartment. The incoming air got to the general passage in front of the compartments first, then was led into the compartments through porous supply ducts arranged above the compartment passage. At the end of this passage each compartment was equipped with a vertical exhaust fan.

The compartments could be heated by warming up the incoming air in the general passage. Additionally a hot-water heating system was installed in the nests of the piglets.

Except for the nests, there were perforated floors (Swing, MIK, Marienhausen, D) in the pens. During the first half of the experiment (dust measurements from 25.05.2000 to 04.04.2001) no straw was used. In the second half (dust measurements since 21.05.2001) the sows received 500 g straw 2 days before the expected date of birth in order to encourage nest building behaviour. In the compartments 1 to 3 it was put into an automatic feeder, in compartment 4 into a trough on the floor. On the second day after the parturition the remaining straw was removed from the pens.

Figure 1:
Locomotion pens, compartment 4

The compartments were managed according to the all in-all out method. The different genotypes kept on the experimental farm were distributed to the compartments at random.

During the whole experimental period feeding was carried out by hand. Initially the troughs were filled directly, but later on automatic feeders were installed each with a capacity of one daily ration.

Measuring the climate inside the stable

The airborne dust concentration was measured in the middle of the compartment passage at a height of approximately 1500 mm. The ambient particulate monitor TEOM 1400a (Rupprecht and Patashnick, Albany, NY) recorded data about the total suspended particulate matter (TSP). It consists of two major components: the sensor unit and the control unit. The control unit contains the data processing hardware, display, flow control components and control electronics for the system. The sensor unit contains the sample inlet and the microbalance (Patashnick and Rupprecht, 1991).

At first the air flows through the sample inlet. In the present investigation a TSP sample inlet was used. This inlet is used extensively in countries speaking German, especially for government mandated ambient particulate monitoring. The TSP inlet does not perform any intentional particulate removal. Instead it prevents unusually large particles that are not representative of suspended particulate matter from

being sampled (Rupprecht and Patashnick, 1993). Since the design flow rate of the TSP inlet is 1 $m^3 h^{-1}$ the monitor was fitted with a flow splitter. According to Rupprecht and Patashnick (1993) it divides the total flow isokinetically into a main flow that proceeds to the mass transducer and a bypass flow. In the present study the main flow was set to 0.18 $m^3 h^{-1}$.

After passing the flow splitter the main flow is guided through a tapered tube which is working as a microbalance. The tapered tube is maintained in oscillation by the feedback electronics. The frequency of this oscillation is measured, the output is evaluated by a microprocessor. The frequency changes as particles land on the filter at the free end of the tube. The relationship between frequency and mass loading on the filter is readily obtained (Patashnick and Rupprecht, 1991).

Since there was only one measuring instrument available, no simultaneous measurements in different compartments were possible. Each measuring period within a compartment consisted of several days in a row. The frequency of the dust measurements was 0.5 Hz.

Additionally, the air temperature and the relative humidity were measured continually in the compartments.

The exhaust air flow rate was recorded daily at 0900 hours by reading the ventilation performance from the ventilating system display.

Data analysis

The statistical analysis of the dust concentration was carried out with the statistic software SAS 8.01. In all cases the dependent variable was the rolling average of the dust mass stored with a frequency of 0.56 mHz.

Initially, data which were not plausible were eliminated. The analysis was restricted to measuring values within the range of 1 to 3500 µg m^{-3}. 3500 approximately equals the mean dust concentration of the whole data set plus 2 standard deviations. Measuring values recorded before the first birth were eliminated as well.

Subsequently, the measuring values were newly numbered. In this numbering, measurement 1 is the first plausible measurement within one measuring period. This is independent of the point in time at which the measuring took place. Measurement 2 is the next plausible measuring value whether there have been eliminated values in between or not.

As a consequence of these data steps the data set comprised 15 301 observations, each representing one plausible measuring value. These values were obtained in 40 different measuring periods (compartments 1 to 4: 11, 9, 10 and 10). Each period was characterized by 69 to 744 continually numbered individual measuring values.

Finally the data were organised in such a way that each measuring period was represented by one observation (proc transpose). Thus, a data set of only 40 observations remained for analysis. In addition to the independent variables each observation consisted of 744 variables. These latter represented the results of the individual measuring values within the particular period. The data analysis was restricted to 672 individual measuring values per measuring period.

This data set was analysed with two different methods following Everitt and Der (1998). Firstly, the means of the particular periods were calculated. With these (n = 40) an analysis of variance was done (proc glm). The second analysis of variance allowed for the fact that there were several, interdependent repeated measurements. However, this analysis was restricted to a relatively small number of repeated measurements, because plausible data were only recorded for a short period of time in some periods. Finally 40 observations with 48 repeated measurements each were analysed (proc glm, repeated *dust* 48).

Results and Discussions

Continuous measurements

Table 1 shows the continuously measured properties of the climate in the stable. The analysis of variance did not reveal a significant influence of the compartments on the dust concentration. The differences between the compartments with locomotion pens were bigger than the difference between locomotion pens and farrowing crates.

Taking the number of weaned piglets per compartment as a criterion for animal activity and furthermore for the quantity of particles (e.g. faeces, skin, hair), this property did not significantly affect the dust concentration either.

Thus, it can not be concluded from the present investigation that, under the given housing conditions, offering nursing sows the possibility to move leads to an increased airborne dust concentration in the compartment. The clear relation between activity and dust concentration reported by Bönsch and Hoy (1996) could not be corroborated. Their study was based on fattening pigs in a deep litter system. The activity of the animals was recorded with the aid of video technique.

Furthermore the analysis of variance did not reveal a significant difference between the periods

with or without straw for nest building. This finding is underlined by a comparison of the figures 2 and 4 on the one hand and the figures 3 and 5 on the other.

Table 1:
Climate in the stable depending on the housing system

		compartment			
		1	2	3	4
D [µg m^{-3}]	avg	301	324	268	349
	±	86	86	55	314
T [°C]	avg	19.3	21.7	21.7	21.5
	±	2.2	2.5	1.8	2.5
rH [%]	avg	61.5	58.3	53.6	58.8
	±	5.7	4.6	6.7	6.4

Average values, avg, and standard deviations, ±, calculated on the basis of the average values by compartment and measuring period (n = 40)
D, Total suspended particulate matter; *T*, Temperature; *rH*, relative humidity

The values of the dust concentration in Table 1 are slightly higher than those found in comparable compartments of the same piggery (Voß, 2001). One possible reason for this is the fact that Voß (2001) mainly carried out her measurements before farrowing.

This explanation is corroborated by the present investigation. The dust concentration measured in the period before farrowing was always lower than in the period until weaning. Whether there is a causal connection with birth could not be settled here. As a consequence of the small number of data the prenatal measuring values were ignored in the analysis (see materials and methods). After the above interpretation of the results of the analysis of variance it does not seem probable that birth and the activities involved result in a considerably higher concentration of airborne dust.

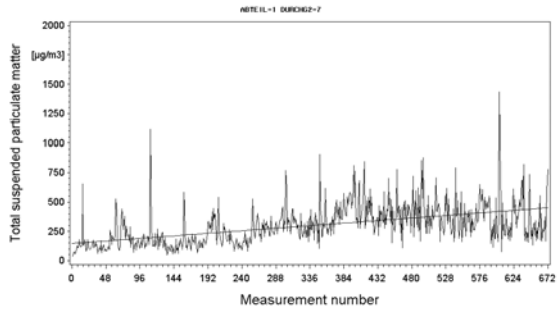

Figure 2:
Course of the dust concentration during one measuring period (compartment 1; no straw was provided to the sows).

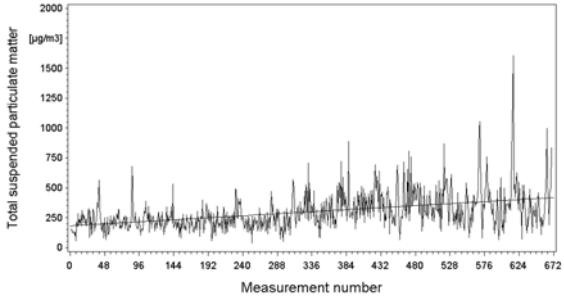

Figure 3:
Course of the dust concentration during one measuring period (compartment 1; straw was provided to allow nest building behaviour).

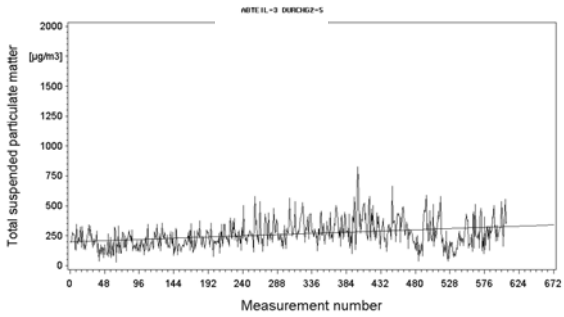

Figure 4:
Course of the dust concentration during one measuring period (compartment 3; no straw was provided to the sows).

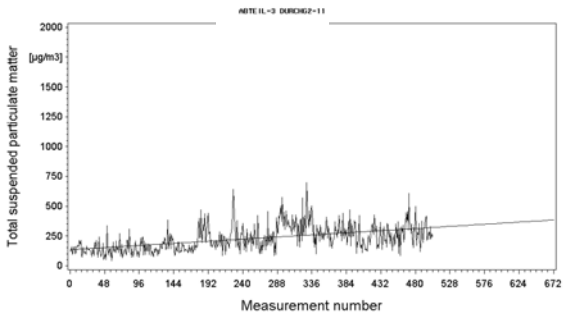

Figure 5:
Course of the dust concentration during one measuring period (compartment 3; straw was provided to allow nest building behaviour).

On the whole, the increased dust concentration is supposed to be caused by a continuous dirt accumulation after housing the sows. A permanent rise of the dust concentration was noticed in 30 of the 40 measuring periods after farrowing. This is shown in figures 2 to 5. No distinct trend was stated in 10 of the measurements.

Besides the dust concentration Table 1 depicts the house temperature and the relative humidity. These

properties prove that the experimental conditions have been homogeneous and within a range characteristic of farrowing houses.

Discontinuous measurements

Since the air flow rate was only recorded once per day simultaneous information about the air flow rate, the dust concentration, the temperature as well as the relative humidity are only available for 18, 26, 21 and 20 single moments representing the climate in the compartments 1, 2, 3 and 4 respectively.

The means of the air flow rate amounted to 1183 ± 571, 1204 ± 592, 2020 ± 349 and 1547 ± 700 $m^3 h^{-1}$ respectively. Figure 6 shows that there was no clear relation between air flow rate and dust concentration.

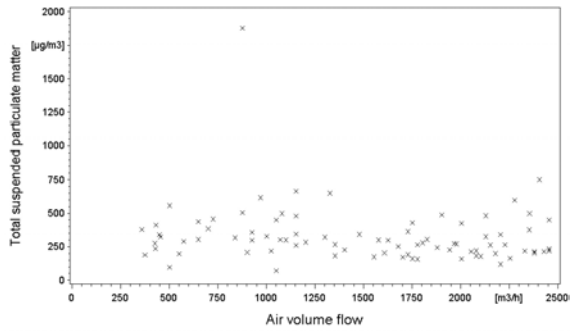

Figure 6:
Relation between total suspended particulate matter and air volume flow (n = 85).

Between the house temperature and the dust concentration no connection was found either, as illustrated in Figure 7.

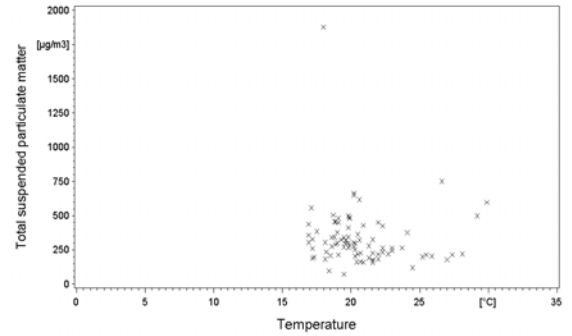

Figure 7:
Relation between total suspended particulate matter and temperature within the compartment (n = 85).

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Particulate matter concentrations in two different buildings for laying hens. A first note

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Abstract

Dust concentration, as Particulate Matter ($PM_{2.5}$ and PM_{10}), was monitored in two different buildings for laying hens in Italy. The first unit, equipped with a traditional system of slurry removal (deep-pit), measured 93x14 m and it housed 60,000 laying hens. The animals were confined in vertical tiered batteries with 5 rows and 6 tiers. The second unit was characterised by a technological system of slurry removal with a low environmental impact, classified as BAT (Best Available Techniques), in which the droppings were dried on ventilated belts. The building measured 80x15 m and it housed 60,000 laying hens. The animals were confined in vertical tiered batteries with 6 rows and 8 tiers. Both the units were ventilated with a negative pressure fan system. The data were collected continuosly every 30 minutes for 34 days in two periods (1st period: August - September; 2 nd period: October - November), using the "Haz-Dust EPAM-5000" which works on-line and it gives the results in real time. The instrument was placed near one of the exaust fans. A data logger recorded temperature and relative humidity continuosly or every 30 minutes.

The data of $PM_{2.5}$ collected during the 1st period were higher for the building with the traditional system of slurry removal than for the building with the BAT technique (0.104 mg/m^3 *vs* 0.027 mg/m^3). The result was the same for the data of PM_{10} (0,38 mg/m^3 *vs* 0.085 mg/m^3).

During the 2 nd period the dust concentration was lower in the first building than in the second one (0.06 mg/m^3 *vs* 0.11 mg/m^3 for $PM_{2,5}$; 0.2 mg/m^3 *vs* 0.48 mg/m^3 for PM_{10}). The result was probably linked to the different values of relative humidity registered during the test. Infact, during the 2 nd period, in the housing using the BAT technology, the average value of relative humidity was 53-55 % while in the other building was higher (64-67 %). As low dust concentrations are associated to higher values of relative humidity, there was a lower efficiency in dust reduction of the BAT technology during the 2 nd period. The very low value of $PM_{2.5}$ in the building with the deep-pit system was linked to the complete emptying of the pit before the data collection.

Keywords: dust, Particulate Matter, BAT, deep-pit, laying hens, ventilated manure belts

1 Introduction

Dust production in poultry buildings is an important problem for both animal and humans health. Dust is made up of different size aerosol particles (with diameters ranging from few microns to hundreds of micron) enabling them to enter the respiratory system. Dust is divided into a "respirable fraction" and an "inhalable fraction", the foremost being the most dangerous since it contains the smallest of the particles (0.5-5 microns) (Van Vicklen *et* al., 1988). If inhaled, they seem to be responsible in humans and animals of allergic reactions and inflammatory processes of the lung because their diameter does not allow them to be expelled (Mølhave *et* al., 2000). These so-called "thin powders", 2.5 micron ($PM_{2.5}$) and 10 micron (PM_{10}), are those more harmful to animals and humans. The environmental control of these two parameters is regulated by italian legislature under the D.Lgs. 626/94, art.33, disciplining security in work places (Ballarini *et* Navarotto, 1995).

In chicken pens, the quantity of dust particles is influenced by a number of factors. Among these, the control of microclimatic parameters is one of the most important (temperature, relative humidity and ventilation rate) (Yoder *et* al., 1988; Reynolds *et* al., 1994; Wathes *et* al., 1998). Dust concentration, infact, is higher in winter than summer (Takai *et* al., 1998). In particular it increases as temperature rises above 15 °C and is maximized at around 21 °C and it decreases after 37 °C (Guarino *et* al., 1999).

Residual feed left on the ground (due to wrong distribution mechanisms as well as the actual quantity of dust present in the feed itself and periodic change of plumage cause an increment of dust levels (White, 1993).

Takai *et* al. (1998) suggested that slurry management and storage as well as the conditions of animal housing represent a source of risk. Literature results prove an increment in dust level as breeding becomes more intensive and in case of floor system *vs* battery system (Ellen *et* al., 2000). Litter also plays an

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important role, as its layers may capture even more dust particles (Takai *et* al., 1998). Even the drying of the droppings collected on the floor is a critical point. In the case of a slatted floor the droppings are at first drained in the pit below the floor and then they are completely removed from the building. Instead, in presence of a full floor, they remain in the building for a long time (Takai *et* al., 1998).

Incremented animal activity related to the presence of light also causes an increase in dust production (Elhussein *et* al., 1999).

2 Objectives

The aims of this study were:

1. to compare the efficiency of two different technologies of slurry removal in reducing dust levels inside the poultry buildings.
2. the evaluation of dust emissions outside the buildings.

In particular the Authors wanted to verify if a technological system of slurry removal with a low environmental impact, called BAT (Best Available Technique), in which the droppings are dried on ventilated belts, is more efficient to reduce dust than the traditional one with deep-pit.

In this phase of the research the follow parameters were collected:

1. the average values of $PM_{2.5}$ and PM_{10} (Particulate Matter Concentrations) inside the buildings.
2. the average values of temperature inside and outside the buildings.
3. the average values of relative humidity inside the buildings.

3 Materials and methods

Description of the building

The experimental study was conducted in Italy in two different buildings for laying hens.

The first unit, organized in two floors, measured 93 x 14 m and it housed 60,000 laying hens (Hy.Lyne W-36).

In this kind of building the ground floor, called “deep-pit”, 2.4 m high, is used for the collection of the droppings. The upper floor is used for the animal housing. The animals were confined in vertical tiered batteries with 5 rows and 6 tiers (figure 1 and figure 2).

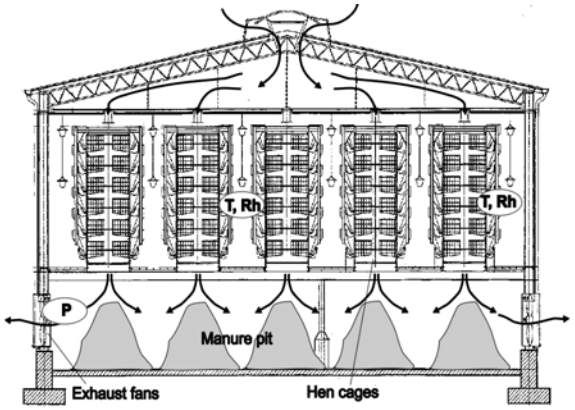

Figure 1:
Cross-section of the laying hen house with the aerated deep-pit for manure storage (traditional system).
Sampling points and measured parameters are specified. (P= particulate Matter; T= temperature; Rh= relative humidity)

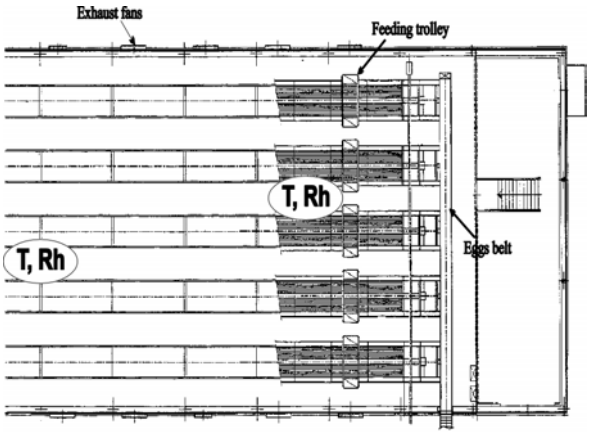

Figure 2:
Layout of the laying hen house with the aerated deep-pit for manure storage (traditional system).
Sampling points and measured parameters are specified. (T= temperature; Rh= relative humidity)

The two floors are communicating because of the absence of flooring in the zone below the cages. The droppings fall on buffle plates under each tier of cages and they’re daily removed by a scraper to the pit under-floor, where they are stored for the whole breeding cycle. The air conditioning inside the building and the drying of the dropping in the pit were controlled by a forced ventilation system. The system was made by 14 axial fans regulated with thermostatic control and placed at level of the ground floor along the longitudinal walls of the structure. An opening on the top of the roof lets air come in and during the passage through the floors it warms up drying the droppings. This technology allows a low energy input

and limited manual working (it is not necessary to move the droppings during a complete breeding cycle). However, the storage of the droppings in the pit for long times get worse the air quality (greater ammonia volatilisation and increment of the dust levels) (Al Homidan *et* al., 1997).

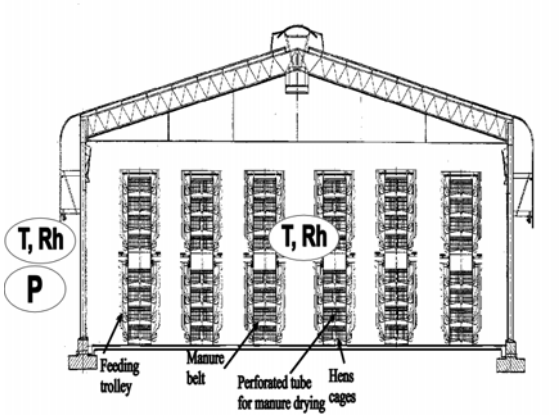

Figure 3:
Cross-section of the laying hen house with the aerated manure belts (BAT system). Sampling points and measured parameters are specified. (P= particulate Matter; T= temperature; Rh= relative humidity)

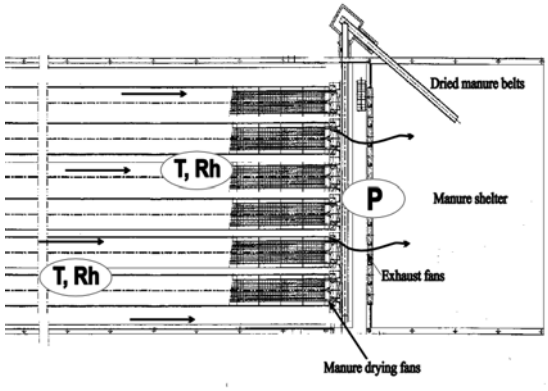

Figure 4:
Layout of the laying hen house with the aerated manure belts (BAT system). Sampling points and measured parameters are specified. (P= particulate Matter; T= temperature; Rh= relative humidity)

The second unit (figure 3 and 4) had dimensions of 80 x 15 m and it housed 60,000 laying hens (Isa Brown) confined in vertical tiered batteries with 6 rows and 8 tiers (figure 3 and figure 4). The manure from the laying hens is collected on a manure belt, (there is one for each tier). Over the belt a perforated tube is placed which blows air over the manure on the belt, drying it. A seven hours (during the night) daily program of ventilation is established and the droppings are weekly removed from belts (every 4-5 days).

For the conditioning of the building a system of longitudinal forced ventilation was adopted (tunnel ventilation). 24 axial fans (16 standards and 6 of emergency) with a maximum air flow rate of 44,000 m^3/h each were used. In Italy, in layer houses, the average minimal winter ventilation rate is about 1.0 m^3/h per hen while the maximum summer ventilation rate is about 10 m^3/h.

Environmental parameters

During the monitoring cycles main microclimatic parameters (inside and outside temperature and relative humidity inside the building) were monitored. These parameters were registered every 1 minute in the ventilated manure belt house and every 30 minutes in the deep-pit system by a computerized system.

Dust levels

The dust levels, as Particulate Matter Concentrations ($PM_{2.5}$ and PM_{10}), were monitored inside the two different buildings. Dust concentration in the indoor air (expressed in mg/m^3) was measured using an instrument of new conception, the "Haz-Dust EPAM-5000", which works on line and it gives the results in real time. The data were collected continuosly every 30 minutes for 34 days in two periods (1st period: August - September; 2 nd period: October – November).

In the building with the deep-pit system the instrument was placed near one of the exhaust fan (figure 1).

In the case of the building with ventilated belts the air sampling was made in correspondence of the fans on the head of the building (figure 3 e figure 4). The measure of the ventilation rate was made measuring the rotational speed of the active fans continuosly. These values were correlated to the flow, determined by anemometric measures.

4. Results and discussion

During the first cycle of sampling we collected 602 data for the $PM_{2,5}$ and for the PM_{10}. Table 1, relative to the first period (August-September 2001), shows the descriptive statistics of the values of $PM_{2.5}$, PM_{10}, temperature and relative humidity. The average values of Particulate Matter were lower for the BAT technology rather than the traditional one (the values of $PM_{2.5}$ were 0.027±0.046 mg/m^3 Vs. 0.104±0,077 mg/m^3; the values of PM_{10} were 0.085±0.04 mg/m^3 Vs. 0,380±0,265 mg/m^3).

Table 1:
Descriptive statistics of the parameters monitored in the two buildings during the first period (August-september 2001)

	Ventilated manure belts			Deep-pit		
	$PM_{2.5}$ (mg/m^3)	**T (°C)**	**Rh (%)**	**$PM_{2.5}$ (mg/m^3)**	**T (°C)**	**Rh (%)**
mean	**0.027**	23.1	55.9	**0.104**	27.1	61.6
minimum	0.004	18.3	36.4	0.018	22	43
maximum	0.502	26.8	73.5	0.280	31.9	76
Std. Dev.	***0.046***	1.5	8.6	***0.077***	1.2	2.7
	Ventilated manure belts			**Deep-pit**		
	PM_{10} (mg/m^3)	**T (°C)**	**Rh (%)**	**PM_{10} (mg/m^3)**	**T (°C)**	**Rh (%)**
mean	0.085	23.5	52.2	**0.380**	27.2	57.5
minimum	0.004	22.6	36.4	0.024	21.4	36
maximum	0.261	26.7	61.2	0.957	33.7	77
Std. Dev.	*0.04*	1.08	3.9	***0.265***	1.4	2.9

Table 2:
Descriptive statistics of the parameters monitored in the two buildings during the second period of sampling (October-November 2001)

	Ventilated manure belts			Deep-pit		
	$PM_{2.5}$ (mg/m^3)	**T (°C)**	**Rh (%)**	**$PM_{2.5}$ (mg/m^3)**	**T (°C)**	**Rh (%)**
mean	**0.11**	23	53.8	**0.06**	22.1	64.5
minimum	0.005	22.4	35.3	0.001	19.5	42
maximum	0.69	24.3	70.7	0.671	26.5	73
Std. Dev.	***0.112***	0.35	4.84	***0.05***	1.9	3.9
	Ventilated manure belts			**Deep-pit**		
	PM_{10} (mg/m^3)	**T (°C)**	**Rh (%)**	**PM_{10} (mg/m^3)**	**T (°C)**	**Rh (%)**
mean	**0.48**	22.8	55.3	**0.2**	21.1	67.2
minimum	0.07	18.3	46.7	0.113	19.5	56
maximum	1.956	23.4	65	0.331	24	73
Std. Dev.	***0.34***	0.6	6.1	***0.04***	1.8	4.9

During the second cycle the data collected were 818. Contrary to our expectations we registered a reversal of the average concentrations of dust for the $PM_{2.5}$ and the PM_{10} during the second cycle (table 2). In fact we found values of $PM_{2,5}$ of 0.11±0.112 mg/m^3 Vs. 0.06 ±0,05 mg/m^3 and values of PM^{10} of 0.48±0.34 mg/m^3 Vs. 0,2±0,04 mg/m^3. These results have been correlated to the unexpected necessity of the farmer to empty the pit before the data collection.

With the future programme of sampling, that will be finished during this spring, we'll make a statistical re-elaboration of the data in order to estimate a possible correlation between dust concentration, microclimatic parameters and management in the two different types of building. Then it will be possible to correlate the same parameters to the ventilation rate in order to extrapolate values of emissions that allow us to begin to model the two different technologies.

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Fine particles and their constituents in Germany - results of denuder filter measurements

Ulrich Dämmgen [1]

Abstract

Ammonia in the atmosphere reacts with atmospheric acids such as nitric acid, hydrogen chloride and sulfuric acid to form ammonium salts, which form a major proportion of the atmospheric aerosol, in particular within $PM_{2.5}$. In contrast to the concentrations of other air pollutants, the concentrations of these so-called secondary aerosols did not decrease proportionally to the decrease of their precursor acids, in particular of SO_2, as their formation is in several ways closely linked to the availability of gaseous ammonia.

The paper presents and discusses measurements of the concentrations of ammonium salts in $PM_{2.5}$ aerosols and their precursor gases made at several locations in northern Germany using denuder filter equipment.

Ammonia and sodium seem to neutralize the acids in the water soluble part of the aerosol totally. The annual cycle of the concentrations of these aerosol constituents can be interpreted as being a function both of the ammonia emissions in spring and air temperature.

Fluxes of nutrients and acidity with aerosols may contribute significantly to the respective overall fluxes into forest ecosystems. Any measures to reduce the areas endangered by acidification and eutrophication will have to make sure that the emission of the precursor gases of aerosols, especially of ammonia, will be reduced. This applies also to measures reducing the concentrations of air-borne particulate matter with respect to human health.

As ammonia originates almost entirely from agriculture, the reduction of the concentrations of aerosols will have effects on agricultural management practices.

Keywords: aerosols, $PM_{2.5}$, ammonia, agriculture

Zusammenfassung

Das Spurengas Ammoniak reagiert mit atmosphärischen Säuren wie Salpetersäure, Chlorwasserstoff oder Schwefelsäure unter Bildung von Ammoniumsalzen, die einen wesentlichen Anteil am atmosphärischen Aerosol, insbesondere an $PM_{2.5}$, bilden.

Im Gegensatz zu den Konzentrationen anderer Luftverunreinigungen sind die Konzentrationen dieser sogenannten Sekundäraerosole nicht in dem Maße zurückgegangen, wie es die Verringerung der Konzentrationen der zu ihrer Entstehung beitragenden Säuren, insbesondere von SO_2, hätte erwarten lassen, da die Entstehung der Sekundäraerosole in mehrfacher Hinsicht mit der Anwesenheit von gasförmigem Ammoniak verknüpft ist.

Der Beitrag stellt Ergebnisse von Messungen zur Bestimmung der Gehalte von Ammoniumsalzen in Aerosolen ($PM_{2.5}$) und ihren Ausgangsverbindungen vor, die an mehreren Orten in Norddeutschland unter Verwendung von Denuder-Filter-Apparaturen gewonnen wurden, und diskutiert sie.

Ammoniak und Natrium neutralisieren praktisch die gesamten wasserlöslichen Säuren im Aerosol. Der Jahresgang der Konzentrationen der genannten Aerosol-Bestandteile lässt sich über die Ammoniak-Emissionen im Frühjahr und die Lufttemperaturen deuten.

Die aus der Abscheidung von Aerosol herrührenden Flüsse von versauernden und eutrophierenden können insbesondere bei Waldökosystemen erheblich zur Gesamtbelastung beitragen. Alle Maßnahmen zur Verringerung der durch Versauerung und Eutrophierung gefährdeten Flächen müssen dafür Sorge tragen, dass die Emissionen der zu ihrer Bildung beitragenden Spurengase, vor allem die von Ammoniak, verringert wird. Dies gilt in gleicher Weise für die Verringerung der Aerosol-Konzentrationen im Hinblick auf die menschliche Gesundheit zu.

Da Ammoniak nahezu ausschließlich aus landwirtschaftlichen Quellen emittiert wird, werden Maßnahmen zur Verringerung der Aerosol-Konzentrationen Auswirkungen auf die landwirtschaftliche Praxis haben.

Schlagwörter: Aerosol, $PM_{2.5}$, Ammoniak, Landwirtschaft

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1 Introduction

Agricultural sources are not only responsible for direct emissions of particles (emission of so-called primary particles), but also for the emission of precursor gases the atmospheric chemistry of which yields secondary particles: ammonia (NH_3) emitted from animal and arable agriculture serves as base in atmospheric reactions with gaseous nitric acid (HNO_3) or hydrogen chloride (HCl) such as

$$NH_3 + HNO_3 \leftrightarrow NH_4NO_3$$

$$NH_3 + HCl \leftrightarrow NH_4Cl$$

or sulfuric acid (H_2SO_4) in aerosol droplets

$$NH_3 + H_2SO_4 \rightarrow NH_4HSO_4$$
$$2\ NH_3 + H_2SO_4 \rightarrow (NH_4)_2SO_4$$
$$3\ NH_3 + 2\ H_2SO_4 \rightarrow (NH_4)_3H(SO_4)_2$$

The first two reactions are fast and reversible. The equilibrium concentrations are within the range of concentrations generally observed in central Europe. The concentration product describing the maximum concentrations of gaseous reactants in the atmosphere is a function of both temperature and humidity: low temperatures and high humidities lead to increased particle concentrations, high temperatures result in thermolysis of particulate NH_4NO_3 and NH_4Cl. At temperatures above 10 °C NH_4Cl is not existing in practice. Reactions between gases yielding solid or liquid particles normally presuppose the existence of condensation nuclei such as soot particles.

Atmospheric H_2SO_4 is a reaction product of sulfur dioxide (SO_2). The three reactions given above are irreversible.

Due to their formation process, secondary particles are normally (more or less) spherically shaped; they have typical aerodynamic diameters between 0.1 and 3 µm. As a consequence, these particles are not subject to sedimentation and hence have long atmospheric half-lives, being capable of long range travelling, i.e. over more than 1000 km (see Garland, 2001, and literature cited therein), and contributing to acidification and eutrophication even in remote areas (Dämmgen and Sutton, 2001). However, if these particles are inhaled, they will pass the human nose, trachea and bronchial tubes and reach the alveoles, where they are deposited. For this reason they have gained specific interest in the past decade and are subject to national and international air pollution control.

Measurements of aerosol size distributions show that about two thirds to three quarters of the total suspended particulate matter (TSP) are in the fraction with aerodynamic diameters below 2.5 µm ($PM_{2.5}$) (c.f. Umweltbundesamt 2001). Although considerable effort has been made during the past decades to reduce the concentrations of particles, concentrations of this fraction of aerosols have not yet lessened proportionnaly, as can be shown by the concentration of S in particles (TSP) (figure 1).

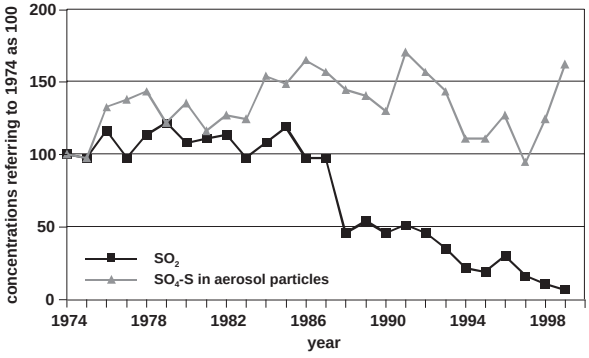

Figure 1:
Effects of air pollution control in Germany: the drastic reduction in SO_2 emissions is not reflected by S concentrations in aerosols (Umweltbundesamt 2001)

The reason for this is the fact that aerosol S concentrations are limited at least to a great extent by the availability of atmospheric NH_3: in non-urban central European atmospheres, $PM_{2.5}$ particle masses are dominated by ammonium salts, their proportion typically being between 50 and 70 % of the total mass. In addition there is crustal material (a few %), so-called elementary or black carbon as well as organic carbon compounds and water. Ammonium salt proportions are highest for particle diameters around 1 µm (c.f. Zinder et al., 1988, ten Brink et al. 1996, Harrison et al. 2002, Hass et al. 2002, Spindler et al. 2002)In the atmosphere, subsequent thermolysis of NH_4NO_3 and HCl combined with chemical reactions of particulate salts with H_2SO_4 as well as dry deposition of reactants and reaction products as outlined in figure 2, should lead to changes in the composition of aerosols: “old” aerosols are likely to be composed of sulfates, whereas “young” aerosols should consist mainly of nitrates.

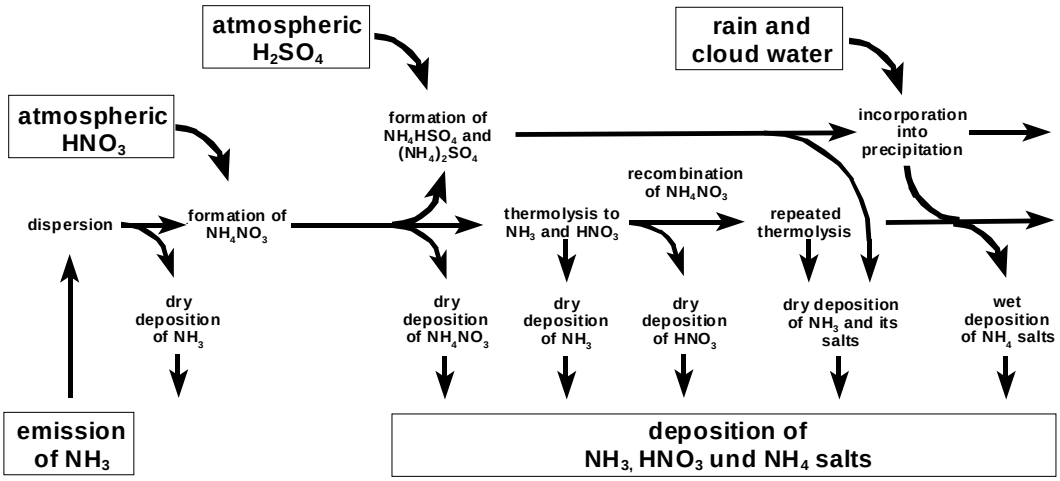

Figure 2:
Emission, atmospheric reactions and deposition processes determining the formation and transformation of aerosol ammonium salts (after Dämmgen & Erisman, 2002)

2 Material and methods

2.1 Sampling of aerosols for constituent determination

Analysis of aerosols is normally achieved by collection on filters with subsequent chemical analysis of the solution obtained when stripping the filter. However, the solutions gained have the same analytes as some gases, which might be collected on the filter at the same time, in particular NH_3, HNO_3 or SO_2, which then fake higher concentrations. At the same time, thermolysis may occur on the filters leading to reduced amounts of NH_4^+, NO_3^- and Cl^-. Therefore the investigation of aerosol constituents has to make use of combined denuder filter samplers. Denuders strip the sampled air from reactive gases (c.f. Dämmgen 2001), so that artefact formation on the filters due to interaction of gases is avoided. The acid gaseous reaction products which stem from thermolysis on an inert (PTFE) filter, are collected on a second (nylon) filter which then allows the back-calculation of the losses on the PTFE-filter.

Combined denuder filter samplers facilitate the determination of the whole set of gases and particles, the concentrations of which are depending on one another as a consequence of chemical transformation, i.e. of gaseous SO_2, HNO_2, HNO_3, HCl, NH_3, and of NO_3^-, SO_4^{2-}, Cl^-, NH_4^+ as well as Na^+ in aerosols. However, operation of batch denuder filter samplers is time consuming, automation is very costly.

For our experiments, we used Kananaskis Air Pollutant Samplers (KAPS, three-stage denuder with PTFE and nylon filter, for details of set-up, operation, analytical procedures involved, and accuracy gained see Zimmerling et al., 1996a, 1997). The samples taken can be analysed for constituents only. Due to potential losses during equilibration in the laboratory, it is (nearly) impossible to obtain filters which can be weighed for the determination of aerosol concentration directly. For such purposes additional sampling of size segregated PM using impactors and filters is necessary.

2.2 Locations of sampling

Since 1991, measurements have been performed of the concentrations of reactive nitrogen and sulfur species in ambient air at various locations

- 1991 to 1993: Rotenkamp near Braunschweig, above grassland
- 1995 to 1998: Müncheberg east of Berlin, above arable land (little animal agriculture)
- 1996 to 1998: Britz/Schorfheide northeast of Berlin, above forest
- 1997 to 1998: Wildbahn/Schwedt in the Oderbruch, in a forest partly influenced by industry and a town
- since 1999 at the FAL site near Braunschweig, above arable land (arable and animal agriculture)

Furthermore two additional sites have been operated at Augustendorf near Friesoythe (Frisia) above forest and Linden near Gießen (Hesse) above grassland since 2001. Results of all campaigns will be used in this paper.

At Melpitz, Saxony, atmospheric aerosol has been investigated using filter techniques since 1993 (Spindler et al. 2002). The results obtained there serve as comparison. No other long-term measurements have been published for Germany (Schaap et al. 2002).

3 Aerosol ($PM_{2.5}$) constituents in northern Germany

3.1 Composition and concentrations

Secondary aerosols are to a large extent composed of ammonium nitrate and sulfates, the ammonium of which stems almost entirely from agriculture. For this reason the total concentration (partial density) of $PM_{2.5}$ ammonium salts will be called the *agricultural aerosol concentration* {$PM_{2.5}$} in this paper. It can be obtained from the partial densities of the constituents analysed.

For 2001, Na concentrations were available almost all of the time. The percentages of the composition are given in Table 1. The anions and cations determined under the assumption that all sulfate is existing as $(NH_4)_2SO_4$ nearly always form neutral salts. A slight surplus of anions, i.e. the potential acidity, could be deduced from the linear regression ($\Sigma\, e_{cat} = 0.97\, e_{an} + 0.004$, $r^2 = 0{,}924$). However, this is also well within the uncertainty caused by sampling and the analytical procedures involved (figure 3).

Table 1:
{$PM_{2.5}$} concentrations at the FAL site, Braunschweig, in 2001, as calculated from water soluble constituents, and proportions of the respective mass contributions

species	unit	Apr - Sep	Oct - Mar	year
{$PM_{2.5}$}	µg m^{-3}	6.8	10.7	8.6
NH_4	%	28	28	28
NO_3	%	24	38	30
SO_4	%	31	19	26
Cl	%	11	12	11
Na	%	5	3	4

Similar measurements had been performed at other locations in different years. A comparison of the results compiled in Table 2 has to be made with due care. However, it shows a reduction in overall concentrations for the one location where samples were taken in different periods: the summer means of $PM_{2.5}$ concentrations at the FAL site obviously decreased considerably. This corresponds with PM_{10} measurements at Melpitz (Spindler et al. 2002).

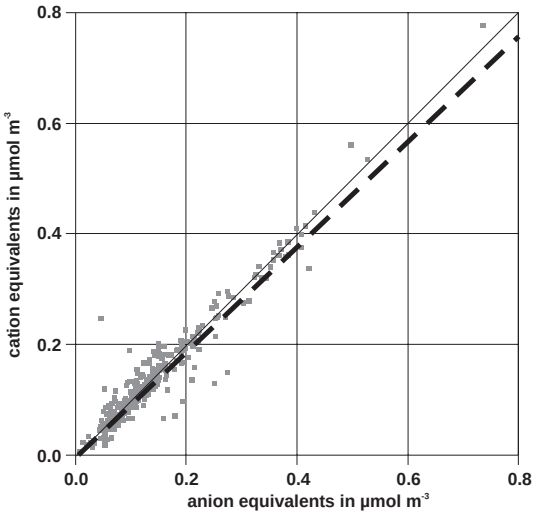

Figure 3:
Anion and cation equivalents in aerosols, denuder filter samples taken at the FAL site, 2001, including NH_4^+, Na^+, NO_3^-, SO_4^{2-}, and Cl^-. The dotted line illustrates the regression mentioned in the text

Table 2:
Mean {$PM_{2.5}$} concentrations in µg m^{-3} at various places in northern Germany, as calculated from water soluble constituents, and proportions of the respective mass contributions. Data from Zimmerling et al., 1996b, Zimmerling et al. 2001, and unpublished data

location	from	to	{$PM_{2.5}$}
Rotenkamp	May 1991	Apr 1993	13.0
	Apr 1993	Aug 1993	10.7
Münche-berg	May 1995	Apr 1998	9.5
Britz	May 1996	Apr 1998	8.7
Wildbahn	Aug 1997	Jul 1998	10.1
FAL	Jan 2000	Dec 2001	8.6
	Apr 2001	Sep 2001	6.8
	Apr 1993	Aug 1993	13.9

At the FAL site, in the (roughly spoken) decade between the measurements in 1993 and 2001, mean concentrations of the precursor gases decreased from about 10 to 2 µg m^{-3} for SO_2, stayed more or less equal for HNO_3 (0.7 µg m^{-3}) or increased considerably for NH_3 (summer 1993: 3.8 µg m^{-3}, summers 2000/2001 6.9 µg m^{-3}).

The parallel measurements in Brandenburg (Münchenberg, Britz and Wildbahn) reveal a difference between the remote forest site at Britz, the agricultural site at Müncheberg and the slightly polluted site at Wildbahn. Although these differences are small, they exceed the resolution of the measurements and are significant. The same order of difference can be seen for the summer period in 1993 when simulta-

neous measurements were performed east (Rotenkamp) and west (FAL) of Braunschweig in areas slightly influenced (Rotenkamp) and considerably influenced (FAL) by animal husbandry. Even in times of increased air pollution, the nearby steel works at Salzgitter and the city of Braunschweig do not emit articles to an extent which can be traced in the rural areas considered (Hartwig-Hanitz et al. 1996).

3.2 Annual and diurnal variation of the concentrations of $PM_{2..5}$ constituents

The annual variation of the species composition of $PM_{2..5}$ with regard to N, S and Cl species is shown in figure 4. The fact that summer concentrations fall below the mean as indicated in Table 2 can clearly be seen. The reduction can obviously be attributed to the decreased proportion of NH_4NO_3.

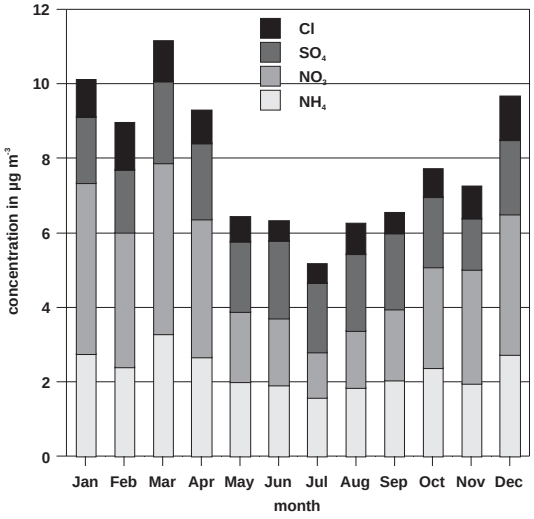

Figure 4:
Annual variation of the concentrations (partial densities) of ammonium, nitrate, sulfate and chloride in $PM_{2.5}$, monthly means for 2000 and 2001, FAL

A comparison of figures 4 and 5 clarifies that the concentrations of the precursor gases are not directly influencing the local concentrations. In particular does the presence of SO_2 in winter not result in a greater proportion of sulfate. Prior to the formation of ammonium sulfates, SO_2 has to be oxidized to form H_2SO_4. This process takes place within droplets; its rate is strongly pH dependent, and "hence the rate of sulfate production tends to be determined by the concentration of ammonia gas" (Choularton and Bower, 2001). Nevertheless, even the comparatively small amount of HNO_3 present leads to effective particle formation.

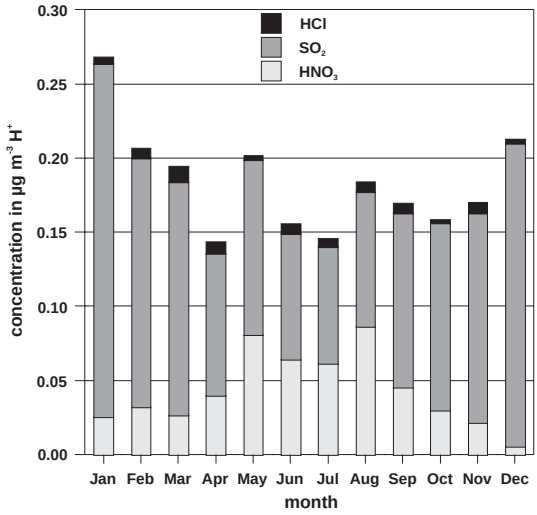

Figure 5:
Annual variation of the concentrations (equivalents) of acid precursor gases for secondary particle formation (HCl, SO_2, HNO_3), monthly means for 2000 and 2001, FAL

Although there is no direct correlation at all between single concentrations of the aerosols and their precursor gases - correlation coefficients between the relevant pairs of species are typically below 0.2, (Zimmerling et al. 2000a) -, the diurnal variation of $\{PM_{2.5}\}$ is obviously related to the variation of the NH_3 concentrations in ambient air. figure 6 indicates that a "mixture" of the annual variations in NH_3 concentrations shown may serve as a key to interpret the annual variation of $\{PM_{2.5}\}$ with a maximum at the time of fertiliser application, again indicating the importance of regional rather than local atmospheric composition.

Obviously concentrations are below average during summer. This can be attributed to minor contributions of NH_4NO_3 to overall $\{PM_{2.5}\}$, most of which can be related to the increased temperatures and thus the increased vapour pressure of NH_4NO_3 (figure 7).

Differences between the agricultural and the forest sites are greatest in March and April showing the influence of regional ammonia sources. This agrees with the interpretation of long-time monitoring results in the USA (Edgerton et al. 1992)

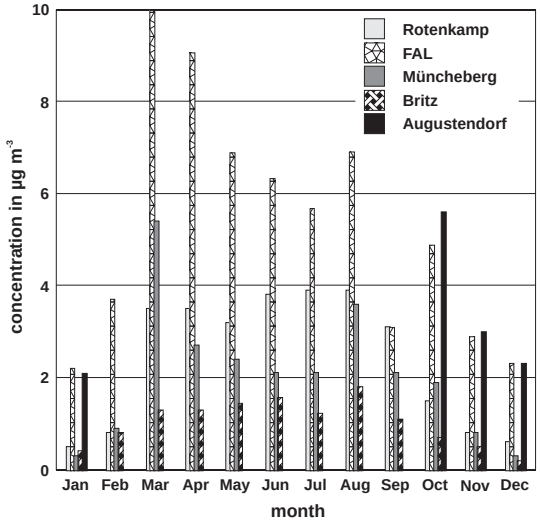

Figure 6:
Annual variation of the concentrations (partial densities) of ammonia at different locations, monthly means for the periods given in Table1, Augustendorf Oct 2001 to Feb. 2002

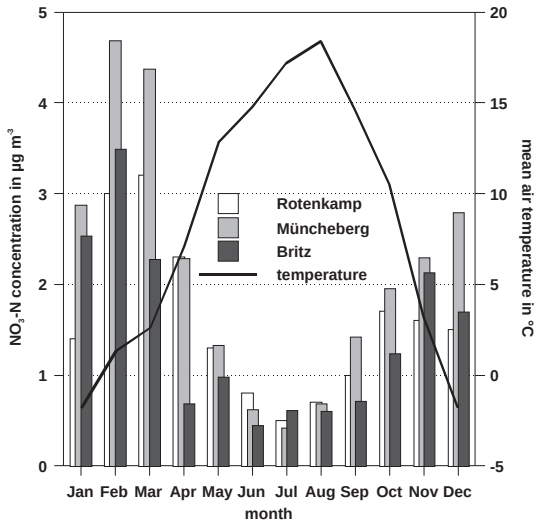

Figure 7:
Annual variation of the concentrations (partial densities) of nitrate N at different locations, monthly means for the periods given in Table 1. Mean air temperature shown is for the Müncheberg region

In contrast to the gases measured, the aerosol constituents exhibit definitely less diurnal variation of their concentrations.

3.3 Concentration profiles and vertical fluxes

Vertical fluxes of gases and aerosols between the atmosphere and a canopy result in vertical concentration gradients, which - with roles reversed -can be related to an atmospheric transport conductance to yield the amount of the vertical flux.

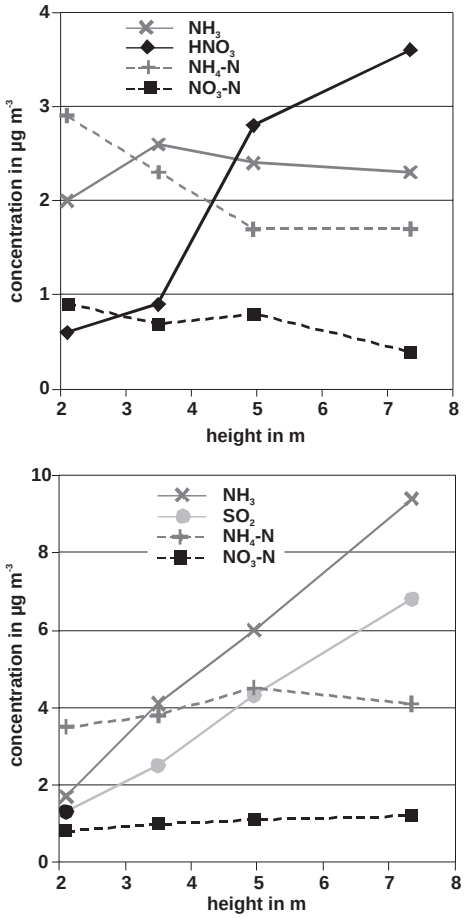

Figure 8:
Vertical gradients of gas and aerosol constituent concentrations, measured at Rotenkamp. Upper half: night-time case study; lower half: daytime case study. Data from Zimmerling (1994)

As can be seen from figure 8, the reactive gases exhibit a strong gradient during night-time, indicating a deposition situation. However, due to the fact that the conductance is very low, no significant fluxes result. The concentration gradients of the aerosol constituents have the same “direction”, but are not distinctive.

The daytime profiles shown in figure 8 look different: the reactive gas HNO_3 has a strong gradient typical for deposition. NH_3, however, reveals an ambiguous pattern, which in combination with those of NH_4^+ and NO_3^- and keeping in mind analytical errors, has to be interpreted in terms of a slight emission from the grassland vegetation and subsequent reaction with HNO_3 causing particle growth near the canopy.

The assessment of vertical fluxes of the aerosol constituents described here shows different behaviour for different types of vegetation. For aerosols and

their constituents, the deposition velocity (i.e. the ratio between flux and concentration of a given species) is strongly dependent on the roughness of the receptor system. Whereas fluxes of aerosol constituents are small for agricultural systems, they contribute significantly to the overall inputs of NH_4-N, NO_3-N and SO_4-S in forests (Hesterberg et al. 1996, Erisman et al. 2001). They may even dominate inputs of eutrophying species into a forest (Zimmerling et al. 2000b).

4 Conclusions

At present, European legislation aims at a reduction of the ecosystems and areas endangered by acidification and eutrophication (cf UN/ECE 2000, IIASA 2002) as well as a reduction in aerosol concentrations (EU 1999). The critical analysis of the composition of aerosols and their depositions clearly shows the importance of the ammonium in aerosols salts for all three fields.

Obviously the availability of atmospheric NH_3 governs the process of particle formation and/or growth. Thus any measure to reduce acidity and nitrogen inputs into natural and semi-natural ecosystems, especially forest ecosystems, will be connected to reductions in NH_3 concentrations. The same applies to reductions of fine particle concentrations in ambient air with regard to human health. As NH_3 originates almost entirely from agriculture, the regulations and recommendations mentioned above will have to be based on a reduction of ammonia emissions, which will have an impact on agricultural production processes, in particular in animal husbandry.

In general, knowledge about aerosols and their constituents in central Europe is thought to be insufficient, which - amongst others - results in poor agreement of measured and modelled concentrations and fluxes of the respective aerosol constituents.

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