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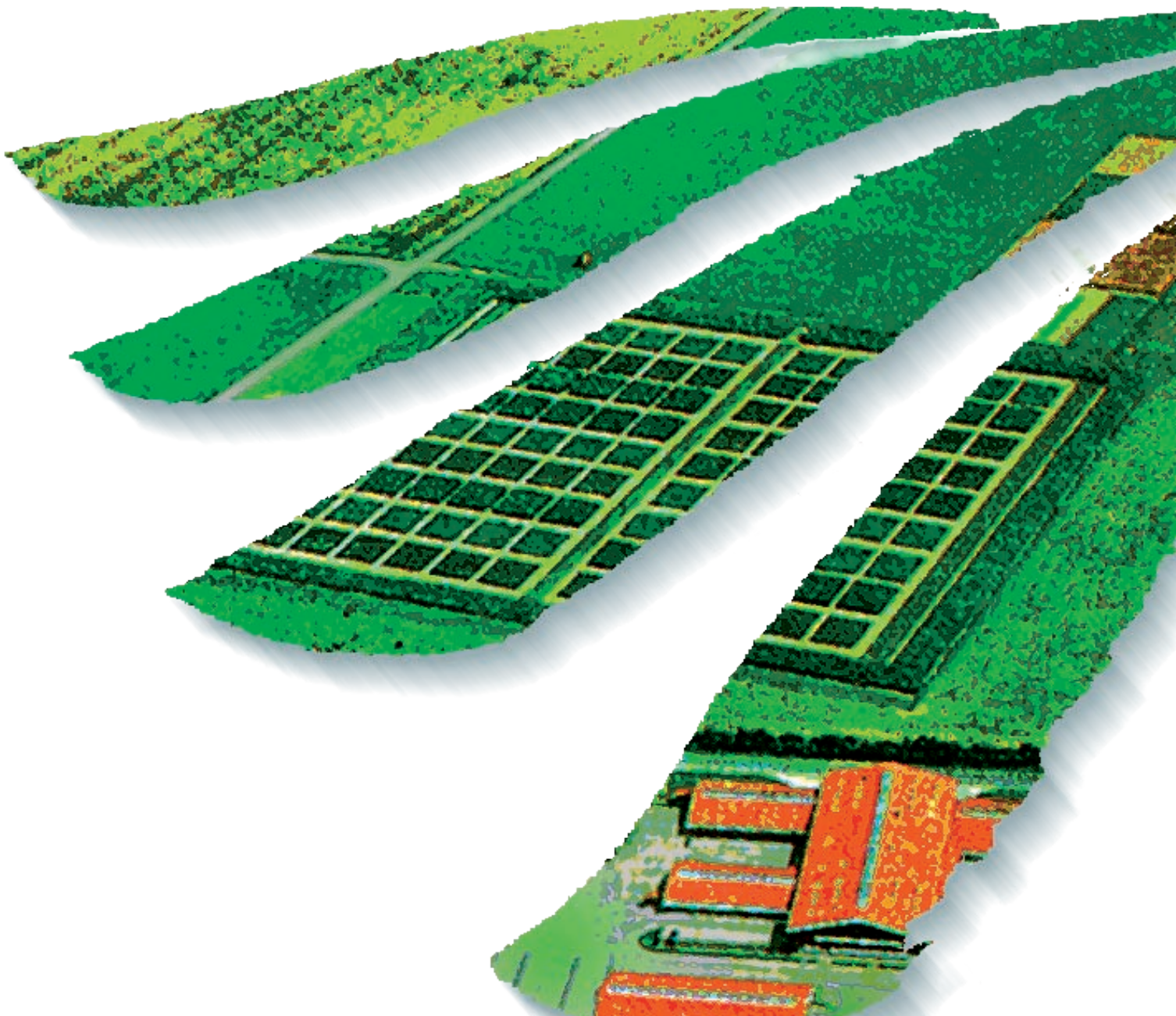
Particulate Matter in and from Agriculture

Torsten Hinz and Karin Tamoschat-Depolt (eds.)



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Particulate Matter in and from Agriculture

edited by
Torsten Hinz and Karin Tamoschat-Depolt

Proceedings of the Conference organized by the
Institut für Technologie und Biosystemtechnik
Bundesforschungsanstalt für Landwirtschaft (FAL)

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Editorial

For the second time the Institute for Technology and Biosystems Engineering of the German Federal Agricultural Research Center (FAL) hosts a conference on particulate matter in and from agriculture. Dealing with research on dust and aerosols in agriculture has a long tradition in this institute starting in the 1950s with experiments concerning cross-flow separation of agricultural goods and the effects of hedge walls and other obstacles on soil erosion. Later on in the 1970s farmer load against dust exposure in arable and stock farming was in the scope of humanisation of labor, followed by considerations of animal protection and environmental relevance.

Currently a wide field of topics is covered by investigations of the PM emissions during soil cultivation, PM emissions in and from livestock enterprises and the emissions caused by the use of bio-fuels in internal combustion engines. The particle size ranges from coarse particles with diameters of hundreds of micrometers down to the ultra fines. Traditional and highly advanced techniques for measuring and modelling are used. Results are available by national and international publications which are quoted in the annual reports of the FAL.

Five years ago at the first conference a state of the art of PM research with agricultural background was given. Data were presented for TSP (total dust) mainly. Some attempts were given concerning the definition of PM 10 and PM 2.5 and the corresponding methods to measure these quantities. For PM emissions from livestock buildings PM 10 and PM 2.5 were calculated by conversion factors based on inhalable dust, which was set to be total dust, and on the respiratory fraction, which was measured according to the convention of Johannisburg. The absolute numbers of the so calculated emission factors may be quite uncertain but the relationships between different species of animals and influences of housekeeping are probably given in a correct way.

Following the new EU regulations about clean air, introducing limit values for concentration of PM in the ambient air, the problem received public awareness. In contrast to e.g. ammonia all forms of fine particles have health impacts. Apparently no threshold for particle concentration can be identified.

Effects including mortality rate are found to be increasing with particle mass concentration in the ambient air. As a result from those findings US EPA decreased its limit value for PM 2.5 to 15 µg/m³, which is much lower than the limit of the EU. WHO is in favour of a value in the near of 5 µg/m³.

But nobody knows exactly how to realize that the limits will be kept. Reduction means are required. Actions must be taken by all stakeholders involved. More political will for reduction is claimed by the authorities. But from the scientific point of view it is necessary first to identify key sources with all relevant parameters in order to turn the right screws.

Agriculture is now identified to be one of the major sources also of PM emissions from livestocks and arable farming. In the Netherlands emissions from animal housing are estimated to contribute 20 % of national PM 10. Ammonia from animal production is one of the key pollutions forming secondary PM 2.5. For an integrated reduction szenario mainly technical end-of-the-pipe measures of separation are recommended although particular other means of reduction are possible.

Arable farming contributes with emissions of soil and produce from the fields caused by natural and machinery induced forces. These emissions were introduced as a major source although its well known that there is a substantial lack of data. The data available are not always comparable since they were collected by applying different measuring techniques and often the description of relevant parameters is missing. But with this concern there are much more gaps to close for air quality in and emissions from agricultural production.

Trying this is one reason for this second conference on PM in and from agriculture. This is in line with the summary of dustconf 2007 in Maastricht, The Netherlands. There it was pointed out that for agriculture the following further actions are needed:

- Funding of international measuring and research programs
- International network for standardisation and harmonisation of measuring techniques and modellers
- Focused integrated study to identify the effectiveness of abatement techniques.

Striking aspects of the conference were questions about:

- Different PM chemical (biological) composition and health effects
- How seasons affect dust emissions, particularly from agriculture and
- How difficult it is to measure PM from livestock housing.

Let us intensively discuss the measurement problems in order to get comparable and reliable data, such that results lead to a well-founded use in emission inventories.

Torsten Hinz

The role of agriculture in the European Commission strategy to reduce air pollution

Z. Klimont[1], F. Wagner[1], M. Amann[1], J. Cofala[1], W. Schöpp[1], Ch. Heyes[1], I. Bertok[1], and W. Asman[1]

Abstract

One of the main EU policy instruments, the Thematic Strategy announced the revision of the Directive on National Emissions Ceilings with new emission ceilings that should lead to the achievement of the agreed objectives with priority given to fine PM. This paper highlights the role of the agricultural sector in the cost-effective emission control strategies for the EU-27. To support the quantitative cost-effectiveness analysis, the GAINS integrated assessment model has been used. In the EU-27, agricultural activities are responsible for the majority of NH_3 emissions and about 4 % of PM2.5; in the future the relative contribution to PM emissions is expected to increase. Implementation of current emission control legislation is estimated to reduce emissions of most pollutants in the EU-27 by 40 - 60 % by 2020. For ammonia, however, emissions are estimated to decline by only 10 %, mainly as a consequence of the expected decline in cattle numbers; taking full account of the Nitrate Directive impact might result in a further 10 % reduction. Also the total control costs are much lower for agriculture than in several other sectors. Although the total potential for technical emission control measures is significantly lower for ammonia than for other pollutants, ammonia offers the largest scope for further emission reductions on top of the measures required by current legislation. In the optimal strategy, a significant reduction of ammonia is expected, which is associated with relatively large costs, representing about 20 % of total additional costs over the current legislation baseline. However, with respect to the total control costs, agriculture remains among the smaller sectors. The analysis performed shows that independent of the environmental objective (excluding an ozone-only case) the emission reductions in agriculture, primarily of NH_3, play an important role in attaining targets specified in the Thematic Strategy and represent about 20 to 40 % of the additional costs of the strategies analyzed. Only in the scenario where full implementation of the Nitrate Directive was assumed is the share of agriculture lower.

Keywords: agriculture, particulate matter, ammonia, National Emission Ceilings, European policy, modelling

Introduction and policy background

A number of studies have demonstrated consistent associations between the concentrations of fine particulate matter (PM) in the air and adverse effects on human health (respiratory symptoms, morbidity and mortality) for concentrations commonly encountered in Europe and North America, e.g., Dockery D. W. et al. 1993; Pope C. A. et al. 2002.

Airborne suspended particulate matter – in the form of primary particles (PM) – are emitted directly into the atmosphere by natural and/or anthropogenic processes, whereas secondary particles are predominantly man-made in origin and are formed in the atmosphere from the oxidation and subsequent reactions of sulphur 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.

Considering the transboundary nature of particulate matter (PM), the Commission of the European Union and the United Nations Economic Commission for Europe's Convention on Long-range Transboundary Air Pollution (UNECE/CLRTAP) are currently developing harmonized international response strategies for a cost-effective control of PM in Europe. While the CLRTAP aims at inclusion of PM-related analysis in the review of the Gothenburg Protocol (UNECE, 1999) in 2008, the European Commission Thematic Strategy on Air Pollution outlined the strategic approach towards cleaner air in Europe (CEC, 2005) and established environmental interim targets for the year 2020 (table 1) within the 'Clean Air for Europe' (CAFE) program (CEC, 2001). As one of the main policy instruments, the Thematic Strategy announced the revision of the Directive on National Emissions Ceilings (2001/81/EC) with new emission ceilings that should lead to the achievement of the agreed interim objectives with priority given to fine particulate matter.

[1] International Institute for Applied Systems Analysis, Laxenburg, Austria

Table 1:
Environmental targets of the EU Thematic Strategy

Effect	Unit of the indicator	Percentage improvement compared to the situation in 2000
Life years lost from particulate matter (YOLLs)	Years of life lost	47 %
Area of forest ecosystems where acid deposition exceeds the critical loads for acidification	km^2	74 %
Area of freshwater ecosystems where acid depotion exceeds the critical loads for acidification	km^2	39 %
Ecosystems area where nitrogen deposition exceeds the critical loads for eutrophication	km^2	43 %
Premature mortality from ozone	Number of cases	10 %
Area of forest ecosystems where ozone concentrations exceed the critical levels for ozone[1)]	km^2	15 %

[1)] This effect has not been explicitly modelled in RAINS. The environmental improvements in the area of forest ecosystems exceeding ozone levels resulting from emission controls that are targeted at the other effect indicators have been determined in an ex-post analysis.

In 2006, the European Commission began the process to develop national ceilings for the emissions of the relevant air pollutants with close involvement of numerous stakeholders including national experts and industrial associations (Amann M. et al. 2006). As a starting point, the analysis developed baseline projections of emissions and air quality impacts to be expected from the envisaged evolution of anthropogenic activities taking into account the impacts of the present legislation on emission controls. Subsequently, a series of reports explored sets of cost-effective measures that achieve the environmental ambition levels of the Thematic Strategy. The reports analyzed potential emission ceilings that emerge from the environmental objectives established and studied the robustness of the identified emission reduction requirements against a range of uncertainties (Amann M. et al. 2007a).

This paper highlights the role of measures in the agricultural sector in the cost-effective emission control strategies, drawing on analyses presented in the NEC reports (Amann M. et al., 2007b) and outcomes of the study on integrated measures in agriculture (Klimont Z. et al. 2007).

Methods

To maximize the cost-effectiveness of emission control strategies, measures for reducing the various precursor emissions of particulate matter (primary and secondary) need to be balanced across all contributing economic sectors, including agriculture, in view of the contributions they make to the various environmental problems. To support the quantitative cost-effectiveness analysis, the GAINS (Greenhouse gas - Air pollution Interactions and Synergies) integrated assessment model has been used to allocate emission control measures across economic sectors.

The GAINS model, which has been developed at the International Institute for Applied Systems Analysis (IIASA), is an integrated assessment model that brings together information on the sources and impacts of air pollutant and greenhouse gas emissions and their interactions. GAINS is an extension of the earlier RAINS (Regional Air Pollution Information and Simulation) model, which addressed air pollution aspects only. GAINS brings together data 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. GAINS addresses air pollution impacts on human health from fine particulate matter and ground-level ozone, vegetation damage caused by ground-level ozone, the acidification of terrestrial and aquatic ecosystems and excess nitrogen deposition to soils, in addition to the mitigation of greenhouse gas emissions. GAINS describes the interrelations between these multiple effects and the range of pollutants (SO_2, NO_x, PM, NMVOC, NH_3, CO_2, CH_4, N_2O, F-gases) that contribute to these effects at the European scale. A detailed description of the air pollution component of the GAINS model can be found in Schöpp W. et al. (1999) and http://www.iiasa.ac.at/rains/review.html; the model is also available in the Internet from http://www.iiasa.ac.at/rains.

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, NMVOC and NH_3. Inter alia, agricultural activities contribute to primary emissions of particulate matter, e.g., livestock housing, arable farming, managing crops, energy use, burning of agricultural waste and unpaved roads. In addition, NH_3 released from agricultural activities constitute an important precursor to the formation of secondary aerosols.

GAINS allows the estimation of emissions from livestock housing, fertilizer application, energy use in agriculture (small stationary combustion and mobile sources)

and, to some extent, from handling of crops. Size-specific PM emission factors were developed, drawing on the results of Takai H. et al. (1998), Louhelainen K. et al. (1987), Donham K. J. et al. (1986 and 1989), ICC &SRI (2000), and Heber A. J. et al. (1988). GAINS estimates country-specific emission rates per animal per year considering the length of the housing periods (for cattle and pigs). Details of the GAINS methodology for the assessment of particulate matter emissions and control costs from agriculture are documented in Klimont Z. et al. (2002). For ammonia the detailed methodology is presented in Klimont Z. and Brink C. (2004).

Historical activity data for agriculture originates from FAO (2006), IFA (2004), and is supplemented by national information collected from national experts. Forecasts up to 2020 have been developed for the review of the National Emission Ceilings Directive of the European Community and are documented in Amann M. et al. (2006 and 2007a).

An integrated assessment needs to link changes in the precursor emissions at the various sources to responses in impact-relevant air quality indicators at a receptor grid cell. Traditionally, this task is accomplished by comprehensive atmospheric chemistry and transport models, which simulate a complex range of chemical and physical reactions. The GAINS integrated assessment analysis relies on the detailed analyses conducted with the Unified EMEP Eulerian model (Simpson O. et al. 2003), and represents the responses in air quality towards changes in emissions as computed by the EMEP model through computationally efficient response surface functions. Such source-receptor relationships have been developed for changes in emissions of SO_2, NO_x, NH_3, VOC and PM2.5 from the 27 Member States of the EU, Croatia, Norway and Switzerland, and five sea areas, describing their impacts for the EU territory with the 50 km × 50 km grid resolution of the geographical projection of the EMEP model (see www.emep.int/grid/index.html).

Quantitative environmental objectives have been established by the European Commission in its Thematic Strategy on Air Pollution for four environmental indicators: Years of life lost (YOLL) from PM2.5 exposure, areas unprotected from eutrophication or acidification, premature deaths from ozone, or jointly on all indicators (see table 1). The optimization module of GAINS has then been used to find cost-optimal control strategies that meet the environmental objectives established in the Thematic Strategy.

Results and discussion

Agricultural activities in the EU-27 are responsible for typically 85 to 90 % of total emissions of ammonia, and they contribute on average about 4 % to primary PM2.5 emissions from anthropogenic sources. For individual countries, the shares of agriculture in PM2.5 vary from less than 1 % to nearly 10 %, due to structural differences in other sources (e.g., the use of solid fuels for home heating, etc.). Because emissions from other sources will decline in the future due to emission control legislation (e.g., tightened emission standards for mobile sources) and ongoing structural changes (e.g., phase-out of solid fuels), agricultural sources will gain in relative importance and will contribute on average up to 6.5 % in the current legislation baseline projection. Typically, about half of the agricultural emissions of PM2.5 originate from open burning of agricultural residue.

To assess the cost-effective scope for further emission reductions, it is necessary to analyze the impacts of the full implementation of current emission control legislation for all pollutants. Figure 1 compares the effects of current legislation with the scope for further technical emission control measures. Full implementation of current emission control legislation is estimated to reduce emissions of most pollutants in the EU-27 by 40 - 60 % by 2020. A notable exception, however, is ammonia where emissions are estimated to decline in the current legislation case by only 10 %, mainly as a consequence of the expected decline in cattle numbers and not due to more stringent emission legislation. While the baseline projection includes likely impacts of the IPPC Directive for farming, it does not take full account of the Nitrate Directive (ND). The latter was analyzed in Klimont Z. et al. (2007), who estimated that by 2020 an additional 10 % of ammonia emissions could be avoided by proper enforcement of the ND (figure 4, left chart).

Although the total potential for technical emission control measures is significantly lower for ammonia than for other pollutants, ammonia offers the largest scope for further emission reductions on top of the measures required by current legislation (figure 1). While for other pollutants more than half of the technical potential forms part of existing law, for ammonia current regulations involve less than one third of the possible measures.

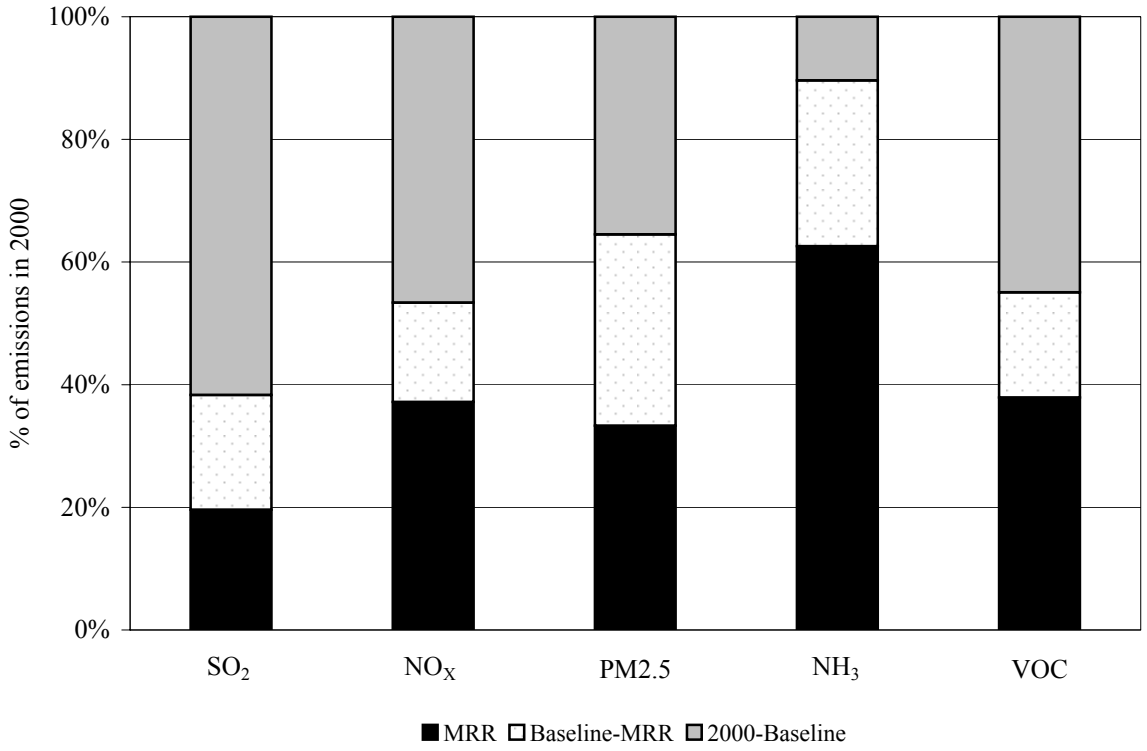

Figure 1:

Scope for further emission reductions in the baseline scenario in 2020, in relation to the emissions in the year 2000.The grey bars indicate emission reductions as a consequence of the full application of current emission control legislation and the white ranges display the potential for further emission reductions that can be achieved with currently available technical emission control measures. The black ranges indicate residual emissions that cannot be reduced with present day emission control technologies.

This paper presents two scenarios that have been explored in the course of the development of the NEC Directive. The first case is the NEC baseline scenario where national perspectives on development of the agricultural sector are considered together with impacts of national and European emission legislation as well as the mid-term review of the EU Common Agricultural Policy (CAP). Its development has been documented in the series of NEC reports, e.g., Amann M. et al. 2007a. Although the principal assumption in the baseline scenario is that the current legislation impacts are included, analysis performed by e.g., Onema O. et al. (2007) shows that stringent interpretation and enforcement of water directives (Nitrate and Water Framework Directives) would require more effective controls and possibly structural changes in agricultural production. Results of the above study were interpreted and implemented in GAINS (Klimont Z. et al. 2007) and this paper presents the potential impacts of such development in the scenario referred to as Baseline + ND (Nitrate directive).

Table 2 shows emissions and total control costs by pollutant for the baseline scenario as well as the optimal multi-effect strategy where TSAP targets (table 1) are met.

Table 2:

Emissions and total costs of the baseline scenario for EU27+Norway (Amann M. et al., 2007b)

	Emissions [Tg/year]			Total costs [billion €/year]		
Pollutant	2000	Baseline - 2020	Baseline - OPT	2000	Baseline - 2020	Baseline - OPT
SO_2	10.3	4.1	2.2	11.0	16.9	19.6
NO_X [1)]	12.5	7.2	5.1	9.8	47.8	51.4
NH_3	4.0	3.6	2.8	1.8	3.4	5.7
PM2.5	1.8	1.2	0.82	8.0	9.3	10.2
NMVOC	11.4	6.4	5.3	0.79	2.3	3.3

[1)] Total transport costs (excluding sulphur-related) included in the NO_x costs

As already shown in figure 1, the agricultural sector emissions do not decline significantly in the baseline and the total costs are far lower than for other pollutants; note that PM2.5 and NMVOC costs exclude the transport sector as all of those costs are associated with NO_x, representing the largest share. In the optimal strategy, significant reductions of ammonia are expected, which are associated with relatively large costs, representing about 20 % total additional costs over the baseline (compare also figure 2 and 3). However, with respect to the total control costs, agriculture remains among the smaller sectors.

Apart from calculating reduction costs of the optimal multi-effect strategy (Joint optimization – figure 2; Baseline – figure 3), we have also assessed the control costs for single effect scenarios (figure 2 and figure 3; Baseline-PM only). For all of the above scenarios the targets were as in table 1. Independent of the environmental objective (excluding the ozone-only case) the agricultural sector emission reductions, primarily of ammonia, play an important role and represent about 20 to 40 % of additional costs in the strategies analyzed. Only in the scenario where full implementation of the Nitrate Directive was assumed (figure 3; Baseline+ND) is the share of agriculture lower. This is, however, largely compensated for by the additional costs in the baseline, as the implementation of ND has been estimated to cost about 0.9 billion € (figure 4, right chart, see the difference in CLE-2020 value).

For the "Baseline+ND" optimal scenario the emission levels of the "Joint optimization" case were assumed as emission ceilings, but the underlying activity data and penetration of abatement measures take into account full implementation of the Nitrate Directive. The Nitrate Directive enforcement has been estimated to bring further reductions of ammonia emissions, about 300 kt NH_3 in the EU-27 baseline by 2020, at an estimated cost of 0.9 billion € (figure 4). The actual costs might be higher when accounting for revenue loss by the farmers who would not be allowed to expand their operations or would have to downscale them, specifically in nitrate vulnerable zones. In the optimal scenario, the overall level of emissions would remain about the same as in the baseline; the total costs to the agricultural sector would be smaller than in the baseline case (figure 4; right panel). This effect is associated with the distribution and level of ammonia emission reductions achieved in the baseline including ND regulation. More details about the implementation and results of this scenario are available from Klimont Z. et al. (2007) and Amann M. et al. (2007b).

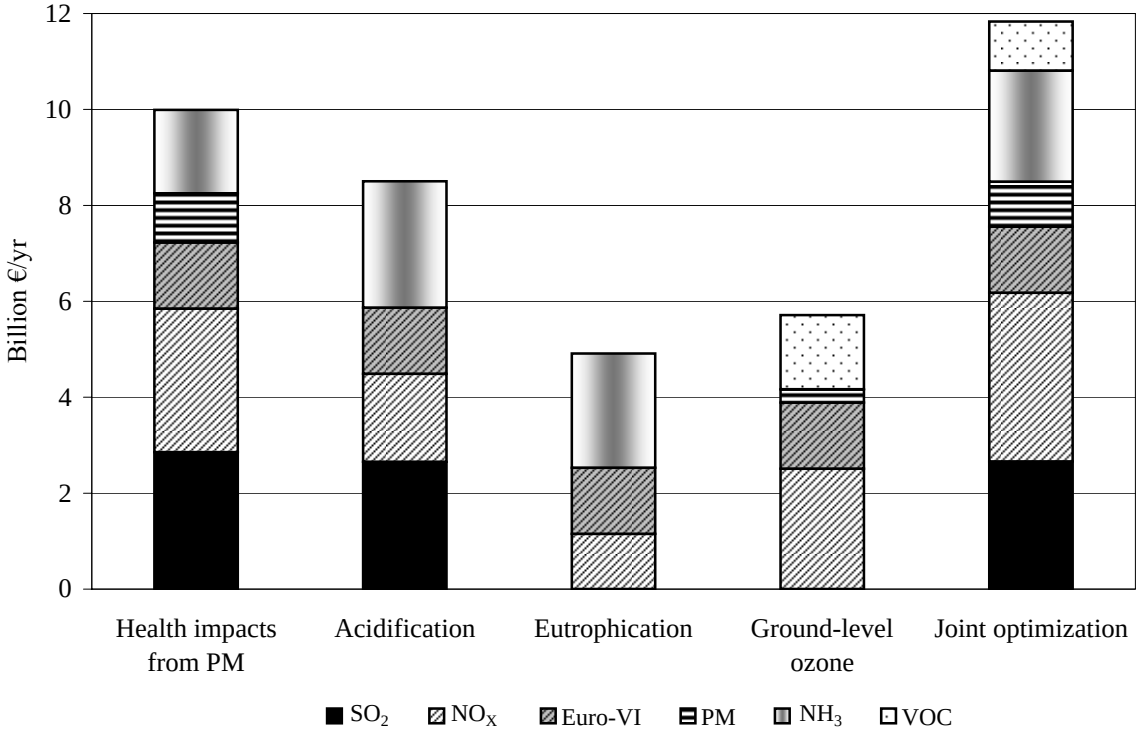

Figure 2:

Emission control costs by pollutant for achieving the four environmental objectives separately compared to the multi-effect (joint) optimization (Where more than one pollutant is reduced by a particular control measure, the allocation of costs by pollutant follows an arbitrary sequence; this may result in otherwise surprising associations between pollutants and impacts, e.g. the apparent influence of PM on ground-level ozone.)

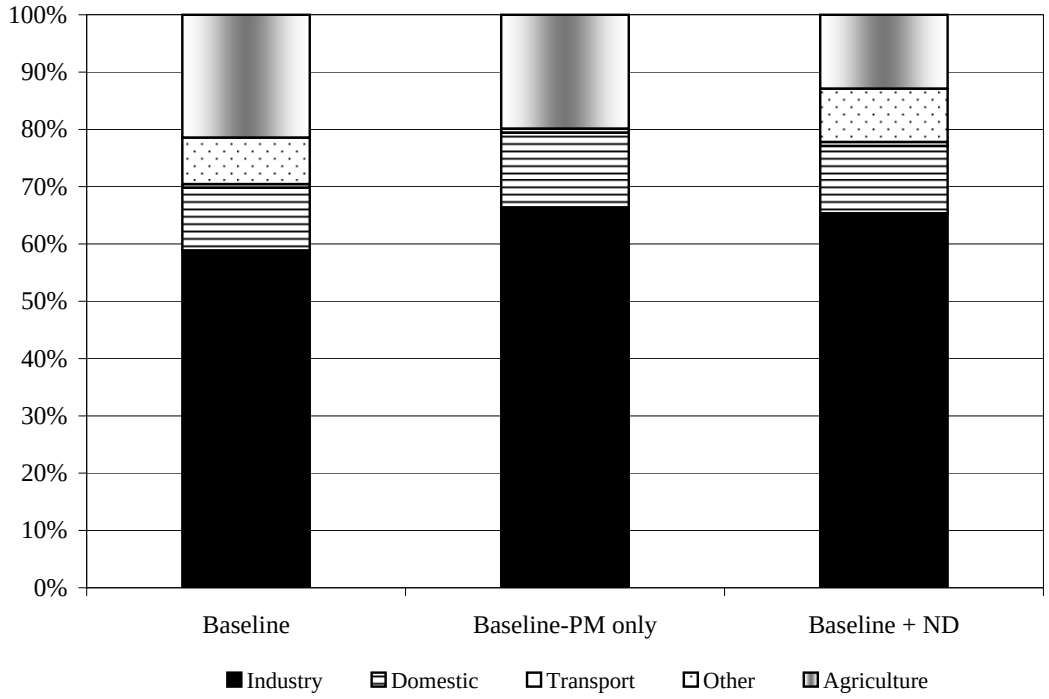

Figure 3:
Distribution of additional (over the current legislation) control costs between sectors for the two optimal multi-effect scenarios (Baseline and Baseline+ND) and the scenario where only the health impacts of PM2.5 are considered (Baseline-PM only).

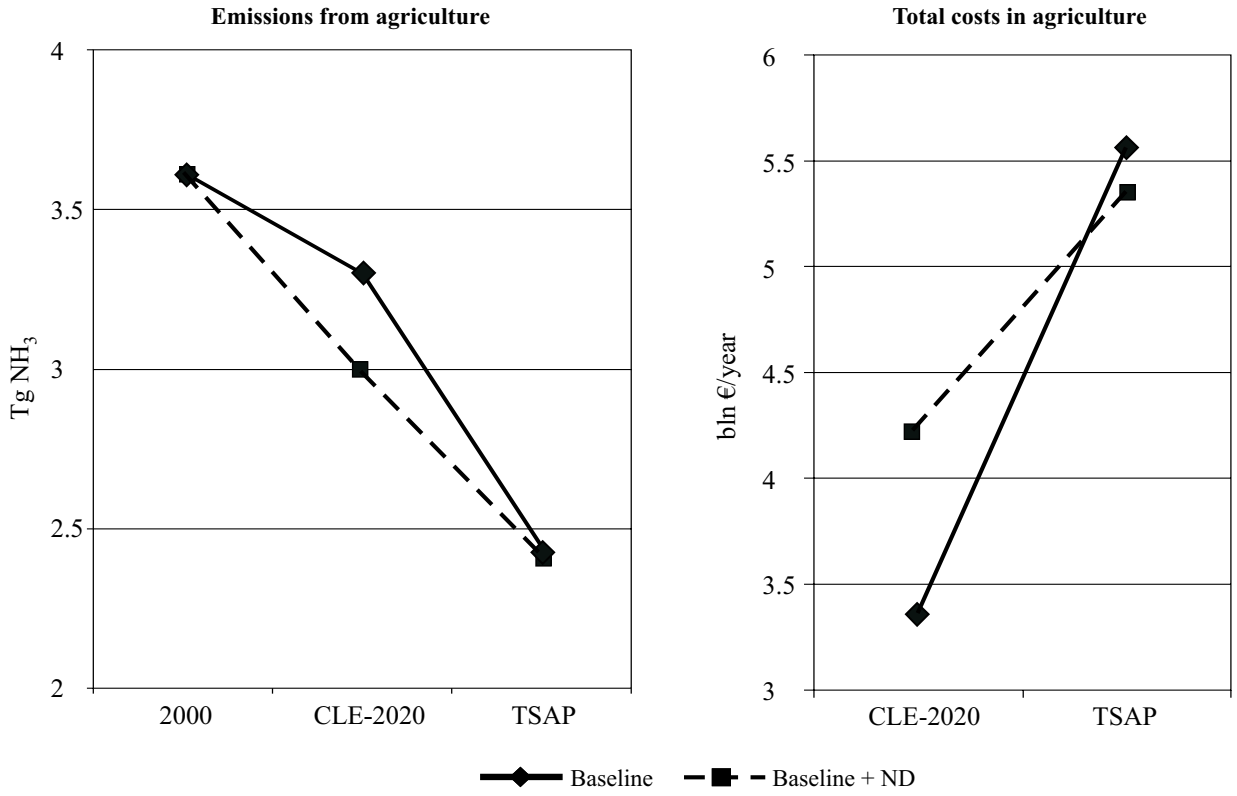

Figure 4:
Expected impact of the Nitrate Directive on agricultural emissions and costs for the year 2020 baseline and optimal scenario with TSAP targets (table 1).

Conclusions

Achieving the health and environmental targets specified in the EU Thematic Strategy will require significant further reductions of emissions of air pollutants in Europe. The analysis performed for the review of the NEC Directive indicates potential for cost-effective reductions of agricultural emissions, primarily ammonia, that contribute to the formation of secondary fine particulate matter and play an important role in excess deposition of nitrogen in sensitive areas. Although total European ammonia emissions are expected to decrease by 2020 by about 10 % compared to 2000, this decline is significantly lower than reductions for other pollutants, so that for secondary aerosols the importance of the contribution from the agricultural sector is expected to grow.

PM emissions from European agriculture are not expected to grow in the next decades. However, in the absence of specific control measures taken in the agricultural sector, its relative contribution to total PM will further increase, from about 4 to nearly 7 %. This is mainly a consequence of stringent emission controls being introduced in other sectors (e.g., stationary energy combustion, mobile sources). Only limited potential for further reductions of primary PM in agriculture has been identified, the main being an effective ban on open burning of agricultural residue that is responsible for about half of the total agricultural emissions of fine PM.

The analysis performed shows that independent of the environmental objective (excluding the ozone-only case) the emission reductions in agriculture, primarily of ammonia, play an important role in attaining targets specified in the Thematic Strategy and represent about 20 to 40 % of additional costs of the analyzed strategies. Only in the scenario where full implementation of the nitrate directive (ND) was assumed is the share of agriculture lower. This is, however, largely compensated for by the additional costs in the baseline associated with the implementation of ND.

Although the calculated emission reductions and additional costs for agriculture are significant, they have to be seen in the perspective of spending already committed by other sectors to control emissions of SO_2, NO_x, PM2.5 and NMVOC. Such comparison shows that the total costs to reduce emissions in agriculture in the analyzed strategies do not exceed 10 % of the total strategy price.

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Trends in emissions and control policies for fine dust particles in Germany

B. Schärer[1]

Abstract

The presentation is on primary, anthropogenic fine particles. We will show the historical trend of fine dust emissions and projections up to 2020 as well as measures and regulations to reduce emissions and influence their trends. Furthermore we will deal with the latest policies in the combat against dust.

Keywords: dust emission, dust projection, policy measures

Introduction

Dust emissions, especially their fine fractions cause unhealthy high concentration in the air in many areas in Germany (By the end of 2006 58 German cities had developed a clean air plan, i.e. that they did not achieve the air quality standard for PM10. The Federal Environment Agency publishes a list of these cities: http:// www.env-it.de/luftdaten/download/public/html/Luftreinhalteplaene/uballl.htm). In addition to the primary dust emissions, the secondary emissions stemming from precursors such as sulphur dioxide, nitrogen oxides, ammonia and volatile organic compounds contribute to elevated levels above the ambient air quality standards. Both, the primary and the secondary particles are due to a wide range of economic, private and natural activities.

The focus of this presentation is on primary, anthropogenic fine particles (i.e. caused by human activities). In this presentation we will show the historical trend of fine dust emissions and projections until 2020 as well as measures and regulations to reduce emissions and influence their trends. Furthermore we will deal with the latest policies in the combat against dust. Ambient air concentrations and their unhealthy effects are not subject of the presentation.

Trend of emissions and regulations for their reduction

Dust is mainly emitted by fuel-burning, which partially serves the generation of energy and the use of vehicles. Other mayor emitting sectors are the handling of bulk materials, production processes, especially the iron & steal works and the mineral industry as well as agricultural activities.

About 40 years ago the two parts of Germany emitted more than 3 million tons of dust per year, with an increasing tendency, even that there were already regulations in place for fitting major new plants with filters. Since that time, and comprehensive and integrated under the 1974 Act of Federal Immission Control (Bundes-Immissionsschutzgesetz - BImSchG), a system of ordinances and technical instructions on emission prevention and control has come into effect and has reversed the trend of growing emissions.

A basic feature of the Federal Immission Control Act is the precautionary principle, which means in practical terms

[1] Federal Environment Agency, Dessau, Germany

that all sources (new and old) must prevent and control emissions according to the state of the art:

1. The establishment and operation of listed plants particularly liable to cause harmful effects on the environment are subject to licensing, relevant smaller installations need type-approval. PM emission control requirements reflecting state of the art technology are laid down in the ordinance on large combustion plants, the Technical Instruction Air (TA Luft) for all other plants subject to licensing and in the ordinance on small combustion plants (<1 MWth).
2. Mobile sources need a type-approval in which the pursuant to EC Directives regulated emissions are tested.

As a consequence of environmental protection policy total dust emission was reduced by over 90 % to about 300 kt in the year 2005. The reduction of dust emissions between 1990 and 2000 of about 1.6 Mt per year was mainly achieved in the new Länder, either by closing old and inefficient power plants and industrial plants, by improving efficiencies of plants and by emission control equipment. Of further significance was the switch from solid fuels to less polluting liquid and gaseous fuels – especially in smaller plants.

Basis for official emission data is the emission data base of the Federal Environment Agency „Central System Emissions – ZSE“ (ZSE, 2005), which is structured according to international reporting guidelines (United Nations Intergovernmental Panel on Climate Change (IPCC): IPCC Guidelines for National Greenhouse Gas Inventories, Reporting Instructions, IPCC-Guidelines http://www.ipcc-nggip.iges.or.jp/public/gl/invsl.htm and of the European Agencie`s Coordination of Information on Air CORINAIR Atmospheric Inventory Guidebook - 2005, CORINAIR-Handbook http://reports.eea.eu.int/EMEPCORINAIR4/en). A break down of the dust emissions by sector is presented in table 1.

Projections for dust emissions

The dust emission inventory of the ZSE was reviewed and adapted to best available scientific knowledge in 2005/6 (UBA, 2007). It now includes all relevant emission sectors. It distinguishes total dust emissions, and according to particle size, PM10 and PM2.5. Sectors new in the emission inventory are abrasion by traffic and the so far hardly elaborated other sources such as cigarette smoke, fireworks and barbequing.

Table 1:
Dust emissions by sectors in Germany, kt

NFR	Quellgruppe	PM	PM10	PM2.5
1 A	**Verbrennung von Brennstoffen**	**108.20**	**70.49**	**52.06**
1 A 1	Energieindustrie	13.06	9.73	8.39
1 A 2	Produzierendes Gewerbe	2.18	2.01	1.64
1 A 3	Transport	56.88	35.09	20.22
1 A 4	Andere Sektoren (Haushalte + Kleinverbrauch)	35.81	23.41	21.54
1 A 5	Andere: Militär	0.27	0.26	0.26
1 B	**Flüchtige Brennstoffemissionen**	**2.45**	**0.71**	**0.35**
1 B 1	Feste Brennstoffe	2.45	0.71	0.35
2	**Industrieprozesse**	**70.97**	**37.58**	**13.00**
2 A	Mineralstoffindustrie	23.53	12.54	4.30
2 B	Chemische Industrie	0.65	0.46	0.29
2 C	Metallproduktion	44.24	22.90	7.34
2 D	Andere Industrieprozesse	2.54	1.68	1.07
4	**Landwirtschaft (PM10)***	**20.52**	**19.86**	**4.39**
4 B	Tierhaltung (Wirtschaftsdünger-Management) (PM10)*	19.33	18.67	4.39
4 D	Bewirtschaftung von Ackerland (PM10)*	1.19	1.19	0.00
6	**Abfallwirtschaft**	**0.01**	**0.01**	**0.01**
6 C	Müllverbrennung: Krematorien	0.01	0.01	0.01
7	Sonstiges	92.80	41.57	17.62
7 A	Schüttgutumschlag	48.35	22.90	4.58
7 B	Sonstiges	44.45	18.66	13.04
	Summe	**294.94**	**170.22**	**87.43**

* Für die Landwirtschaft wurden keine Gesamtstaub-Emissionen berechnet, deshalb werden hier die PM10-Emissionen aufgeführt.

Based on emissions control measures and regulations, that were already in place in 2005, projections to 2020 are shown in figure 1 by sectors and in figure 2 by fractions. The major feature of the future trend of PM-emissions is the substantial decrease, triggered by regulations already in place. The decrease of emissions for all fractions will be quite substantial and will amount to 40 % for PM2.5, 31 % for PM10, and 27 % for total dust.

A breakdown of total emission shows significant differences in the shares of dust fractions of emitting sectors over time.

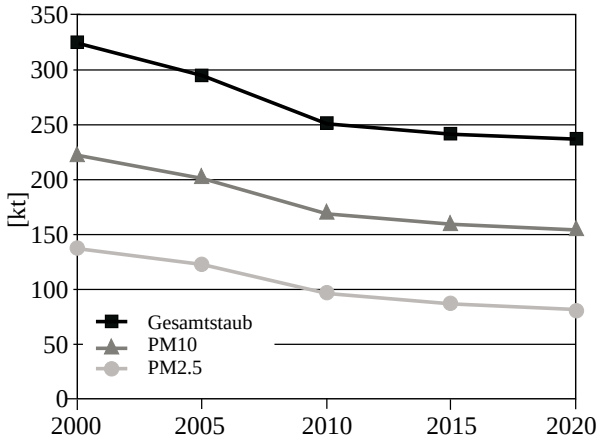

Figure 1:
Primary dust and fine dust emissions in Germany 2000 - 2020

When it comes to total dust, figure 2 shows that emissions are scattered widely over the sectors. On the other hand for PM2.5, which figure 2 does not show, emissions are limited to the sectors road traffic, wood firing, mobile machines, large combustion plants and the ore and steal industry. Agricultural activities also contribute significantly to dust emissions. At the moment agriculture produces 10 % of primary PM10 with a slightly lower share for PM2.5.

For wood burning the emissions depend on the assumptions on the future use of wood. The projections provided in the figures do not include the latest policies and trends of the use of bio fuels. Therefore the emissions for wood firing are underestimated as the use of wood has increased substantially and will grow further. This would lead to a substantial increase of dust emissions without further reduction measures. To cope with this development the Ordinance for small firing installations is under revision with stringent emission limit values.

Outlook

Reducing emissions of fine particulate matter is, in principle, possible for all sources other than natural. With a view to the need for further emission reductions, the German Federal Environment Agency has examined additional measures and their realisation potential. This led to the identification of measures for coal-fired power plants, small-scale wood burning, road and other transport as well as for non-road mobile machinery. The combination of all

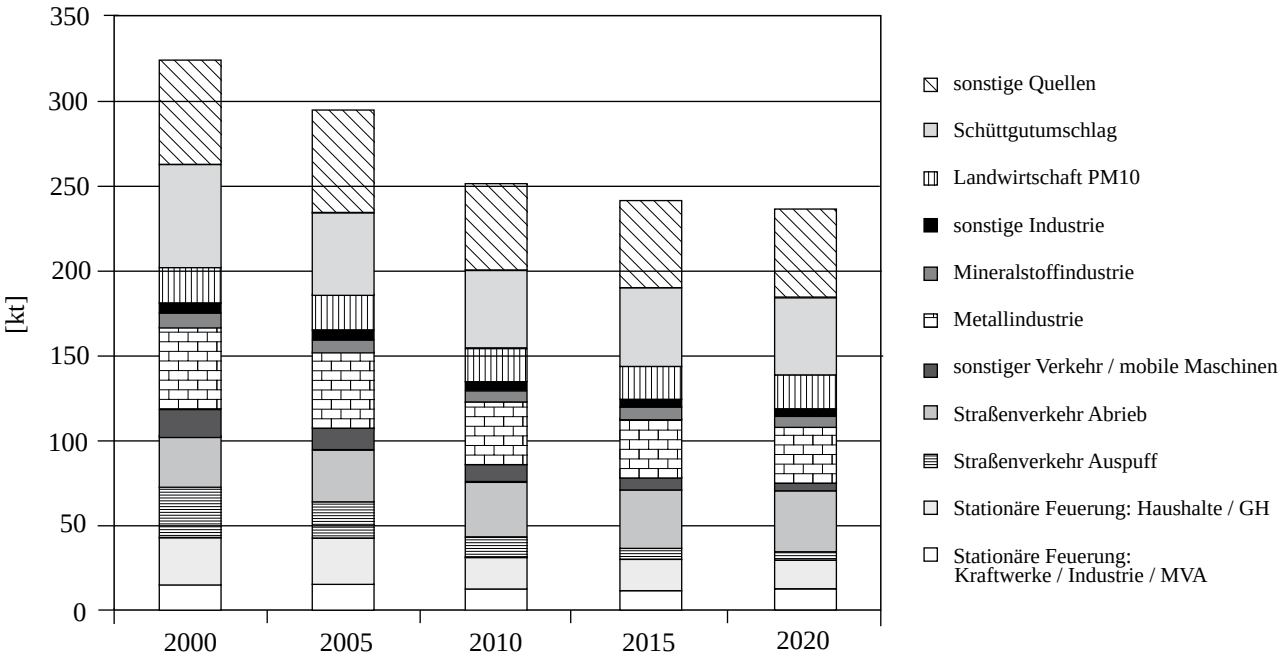

Figure 2:
Breakdown in sectors of the total primary dust and trend of emissions in Germany 2000-2020

measures analysed can reduce emissions of PM2.5 in the reference scenario by 3.3, of PM10 by 2.0 % and of total particulate matter by 1.6 % by 2020.

Due to long-range transboundary transport of particulate matter and precursor substances, national regulations are not sufficient. As part of its air pollution control policy, the EU has developed a strategy to reduce pollution by particulate matter under its Clean Air for Europe Programme and has started revisions of the First Daughter Directive on Air Quality and the NEC Directive. In 2008 the European Commission is expected to propose an update to the NEC Directive up to the year 2020. In addition to defining new national emission ceilings for the substances covered by the present Directive, it is considering introducing national emission ceilings for fine particulate matter (PM2.5).

The United Nations Economic Commission for Europe (UNECE), whose Member States cover a geographical area even larger than that of the EU, has also put pollution by particulate matter on its agenda and will update the so-called Multi-component Protocol to the UNECE Convention on Long-range Transboundary Air Pollution.

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Particulate matter emissions from arable production - a guide for UNECE emission inventories

K. W. van der Hoek[1] and T. Hinz[2]

Abstract

Within arable production we distinguish the following subsequent stages: soil cultivation (includes all working steps treating the soil e.g. ploughing, harrowing, and seeding), harvesting and post harvest treatments at farm scale (like unloading, cleaning and drying crops).

For the arable crops wheat, rye, barley and oat, a first estimate of the emission factor is 3 – 5 kg PM10/ha. The actual and local emission factors are dependent on fixed parameters like soil properties (sand, loess, silt fraction) and variable parameters like dry or moist soil.

It is assumed that a part of the emitted PM10 is deposited in the field and will not leave the field. The part that leaves the field is considered to be inventory relevant. The in-field reduction percentage is dependent on atmospheric stability and wind speed.

Keywords: PM10, arable farming, tillage, emission rates, in-field deposition

Human health aspects

Thomas Jefferson's Declaration in 1776 states that 'Cultivators of the earth are the most valuable citizens. They are the most vigorous, the most independent, the most virtuous, and they are tied to their country and wedded to its liberty and interests by the most lasting bonds'. Unfortunately, the myth of the robust, reliably healthy farmer does not correspond with the realities of agricultural life. Respiratory diseases associated with agriculture were one of the first-recognized occupational hazards. As early as 1555 there was a warning about the dangers of inhaling grain dusts, but it has only been in the 20th century that respiratory hazards in agriculture were studied and documented (ATS, 1998).

Respiratory health hazards in agriculture are documented in full detail in a conference report of the American Thoracic Society, and in a review article (ATS, 1998; Eduard W. 1997). Both studies comprise animal agriculture as well as arable production. Recently a review article has been published devoted to soil as a source of dust and the associated human health aspects (Smith J. L. and Lee K. 2003).

Focus on arable production

This paper is concerned with the emission rates of particulate matter during arable production, storage and handling products while producing food and non food plants and fruits. Not included in the paper are emissions from movement on unpaved roads, from the consumption of fuels and emissions due to the input of pesticides. Also pollens which are mainly larger than the particle sizes concerned in this paper, are not included. Wind blown particles from cultivated soils not arising directly from field operations will be considered as natural emissions. These emissions, often called wind erosion, are very variable as well in size as in frequency.

Arable production in more detail

Different types of soil cultivation, harvesting, and the application of mineral fertilizer are responsible for particulate matter emissions from the fields. Soil cultivation includes all working steps treating the soil e.g. ploughing, harrowing, and seeding. Post harvest treatments like unloading,

[1] National Institute for Public Health and the Environment, Bilthoven, The Netherlands

[2] Federal Agricultural Research Centre, Institute for Technology and Biosystems Engineering, Braunschweig, Germany

cleaning and drying crops are only taken into account if they take place on farm level. Farm level includes all operations on the farm until the produce leaves the farm.

The main sources of particulate matter emissions are caused by combine harvesting and soil cultivation and their magnitude is within the range of more than 80 % of total PM10 emissions from arable production.

Emission of particulate matter in arable production

Emissions of particulate matter in arable production occur from different sources and at different times. Sources are operations on the fields and the farms. In chronological order follow from spring to autumn soil cultivation, harvesting, post harvesting treatments and again soil cultivation (figure 1 and 2).

Figure 2:
Soil cultivation

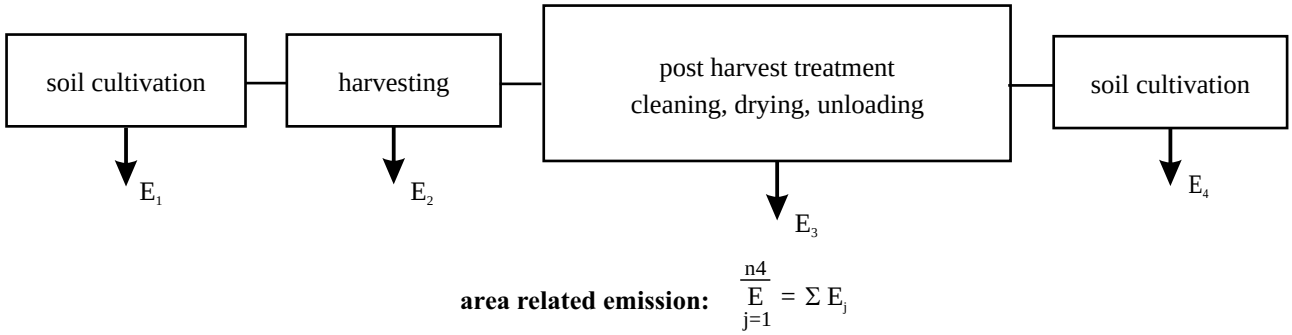

Figure 1:
Scheme of particulate matter sources in arable production

- Mass flows of emitted particles are governed by a large number of parameters:
- the produce, type of crop, fruit, vegetable
- the physical properties of the particles depending on their origin
- origin of the particles: soil, plant, machinery
- soil composition (sand, loess, silt fraction)
- meteorological conditions of soil and/or produce before and during the operation (wind speed, temperature, rain fall, humidity)
- type of operation (harrowing, discing, cultivating, ploughing)
- parameters of the machinery (working speed, working capacity, working surface).

Particle emissions from arable production may be related to the cultivated area of each produce.

$$E_{10} = \sum_{=1}^{n} EF_{10} \cdot A \cdot n$$

E_{10}	emission of PM10 in kg/year
EF_{10}	emission factor in kg/ha
A	annual treated area in ha
n	annual repetitions of treatment

The emission factors of post harvest operations are generally given related to the amount of handled mass. These emissions factors can be converted to area related factors by multiplying with the averaged annual yield:

$$EF_{10} = EF_{10m} * Y$$

EF_{10}	emission factor in kg/ha
EF_{10m}	emission factor in kg/ton
Y	averaged annual yield in ton/ha

The emissions during arable production follow the seasons and are given in table 1.

Table 1:
Time table for considerable working steps in arable production

	Working step, time table			
Crop	Soil cultivation/seeding	Harvesting	Cleaning	Drying
Wheat	March/October	July/August	July/August	July/August
Rye	March/October	August	August	August
Barley	March/September	June/July	June/July	June/July
Oat	March	August	August	August

Definitions of PM10, PM2.5 and TSP

There are different definitions for particle fractions, but all of them define penetration curves of virtual separators. PM10 and PM2.5 origin from US EPA defined for environmental purpose. ISO gives health related definitions which are considering the pathway into the human breathing apparatus.

Figure 3 shows these different curves. Differences are obvious for PM10 and the thoracic fraction which correspond with it by the same cut off at 10 µm. PM10 do not consider particles larger than 15 µm while thoracic reaches up to 40 µm.

particulate matter. This must be considered for PM10 and thoracic fraction if the emissions include a high portion of particles with size between 15 µm and 40 µm. Practical measuring equipment will often follow the ISO definition. Definitions for PM2.5 and the respirable fraction (risk group) are consistent.

TSP means total suspended particles and it is mainly used in ambient air for sizes below 57 µm. From emission point of view TSP means more or less total dust considering all sizes up to the largest particles which size depends on the origin of the dust.

Dust particles should be limited to sizes not larger than 500 µm (aerodynamic diameter).

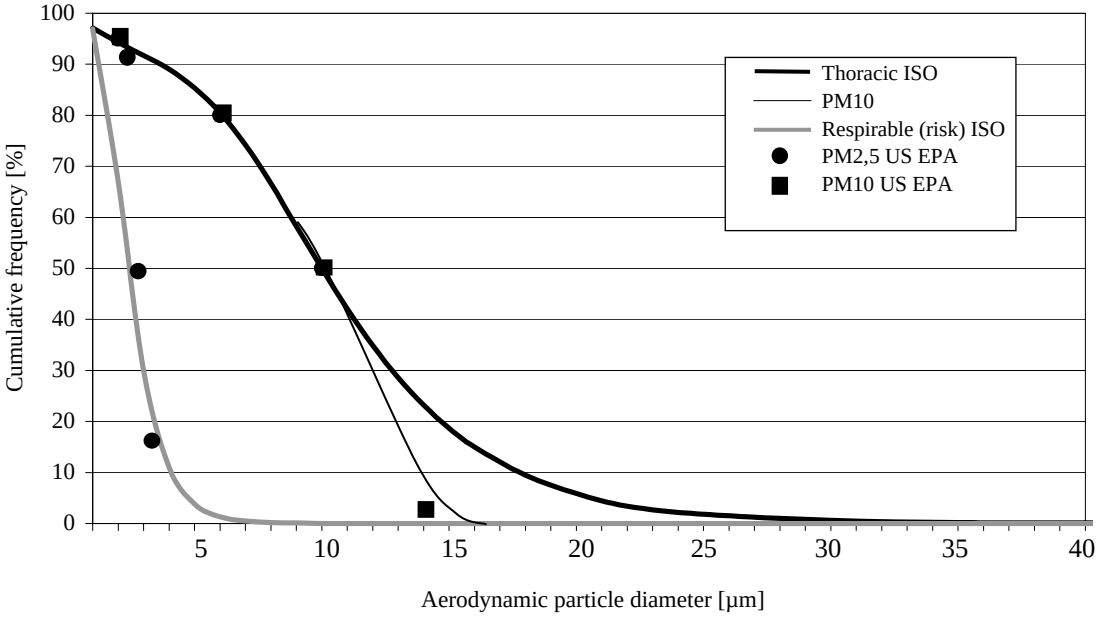

Figure 3:
Patterns of different particle fractions

The presented curves describe virtual particle separators simulating the corresponding parts of the breathing tract. They are characterized by their shape and by the 50 % value of separation and penetration the so called cut off diameter. Samplers with same cut off diameters but different shaped penetration curves will collect different fractions of

Compilation of emission factors

There are different methods for establishing emission factors for arable production.

- Direct measurements of the particulate matter emission flows of tractors and implements. From these machinery

related data of the potential strength of a source, field related emission factors must be calculated.
- Indirect estimation of source strength using concentration measurements carried out machinery bound on the drivers place and models of a layer or a plume on the treated area to get the connection with a balance volume or a volume flow rate concerned.
- Measurements of particulate matter concentrations at the border of a field fitted to an inverse computing model of dispersion.

There is a lot of information on measurements available form California (Baker J. B. et al. 2005, Clausnitzer H. and Singer M. J. 1996, 1997, Holmén B. A. et al. 2000, 2001), Germany (Batel W. 1975, 1976, 1979, Goossens D. et al. 2001, Hinz T. and Funk R. 2007, Oettl D. et al. 2005) and Belgium (Bogman P. et al. 2007).

For soil cultivation the following PM10 emission factors are found:
0.1 kg/ha, RAINS (Klimont T. et al. 2002)
0.06 - 0.3 kg/ha (MAFF 2000, Wathes C. M. et al. 2002)
0.28 - 0.48 kg/ha (Hinz T. et al. 2002).

In table 3 an averaged field emission factor of 0.25 kg/ha is used.

Measurements in California are much higher, 4.2 - 5.2 kg/ha (WRAP, 2006). The reason is probably the climatic and soil conditions with higher temperature and lower humidity. This assumption is supported by measurements done in Brandenburg, Germany under the 2006 hot and dry conditions, resulting in a dry soil and emission values one order of magnitude higher than in former years (table 2).

Table 2:
Emission factors for soil operations (Oettl D. et al. 2005, Hinz T. and Funk R. 2007).

Emission factors for PM10, PM2.5 and PM1 for field operations			
	PM10 kg/ha	PM2.5 kg/ha	PM1 kg/ha
Harrowing	0.82	0.29	<1
Discing	1.37	0.12	0.03
Cultivating	1.86	0.06	0.02
Ploughing, dry soil	10.5	1.3	0.1
Idem, moist soil	1.2	0.05	0.01

For combine harvesting the following PM10 emission factors are found:
4.1 - 6.9 kg/ha, parameter cereal, cereals humidity during harvesting (Batel W. 1976)
3.3 - 5.8 kg/ha (WRAP, 2006).

PM10 emissions from arable production originate on the spots where the tractors and the machinery operate. We have to distinguish between these emissions and the emissions leaving the agricultural field. The latter emissions are much lower by self cleaning effects of the dust plumes by settling and by washing out of fine particles by large particles. This will be discussed in the next section.

Validation of emission factors

From recent US research it is known that regional air quality models overestimate the contribution of PM10 emissions from unpaved roads to the ambient PM10 concentration. A 'dividing by four of the emission' approach is often used as correction term. This overestimation is explained by rapid near source deposition resulting in a smaller transportable fraction to far-field places (Dong Y. et al. 2004). Important parameters for local deposition are land cover type, atmospheric stability and wind speed. Local deposition is reduced under unstable atmospheric conditions and high wind speeds (Etyemezian V. et al. 2003, Pace T. G. 2005, Hagen L. J. et al. 2007).

The same mechanisms hold for PM10 emissions from arable production. It is assumed that only a part of the emitted PM10 leaves the field. Only this part is considered to be inventory relevant. In table 3 two situations are presented: one with 50 % of the original PM10 emissions leaving the field and one with 10 % leaving the field.

Draft emission factors for the UNECE Emission Inventory Guidebook

Based on the cited literature in the section on compiled emission factors we constructed table 3. The PM10 emission factors are a first estimate and the final emission factors will be dependent on the following factors:
- Fixed parameters like soil properties (sand, loess, silt fraction)
- Variable parameters like dry or moist soil
- In-field reduction percentage (unstable/stable atmospheric conditions, wind speed).

Table 3:
Matrix of field emission factors for considerable working steps and totals for crops, as presented as draft in the expert group of the UNECE Emission Inventory Guidebook in Thessaloniki, 2006.
NB The working steps EF are measured on the spot; the amount of PM10 leaving the field is given for 2 situations: 50 % and 90 % reduction by settlement in the near-field.

	Working step EF_{10} kg/ha				EF_{10} kg/ha		
Crop	Soil cultivation	Harvesting	Cleaning	Drying	No reduction	50 % red	90 % red
Wheat	0.25	2.7	0.19	0.56	3.70	1.85	0.37
Rye	0.25	2.0	0.16	0.37	2.78	1.39	0.28
Barley	0.25	2.3	0.16	0.43	3.14	1.57	0.31
Oat	0.25	3.4	0.25	0.66	4.56	2.28	0.46

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Control of PM emission from livestock farming installations in Germany

E. Grimm[1]

Abstract

In order to control the emission of particulate matter and to prevent harmful effects on people living in the vicinity of livestock installations in Germany, potential environmental effects are assessed during the licensing procedures. Assessment is based on the regulations of the Technical Instructions on Air Quality Control (TA Luft), which determines limit values for the immission of fine particles (PM10). These limit values are cited from the European Air Quality Daughter Directive.

For reasons of commensurability a detailed assessment whether the limit values are not exceeded is only required under certain conditions. Assessment is not necessary, if people do not permanently live in the vicinity of an installation and if the so-called bagatelle-emission rate, which is equivalent to a livestock of about e.g. 2,150 fattening pigs and 74,500 laying hens is not exceeded. It is also not required 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 (< 1.2 µg/m³) and additional measures for emission abatement are taken.

In contrast to the regulations of the TA Luft to control the immission load, regulations on emission limitation for total dust in general will not affect livestock installations.

Practical experience reveals, that there is a lack of sound data relating to the emission of PM from different production and housing systems and that there is a need to establish and to harmonise special measurement protocols for their determination.

Keywords: livestock installation, particulate matter, emission data, immission limit values

1 Introduction

Agriculture, namely livestock installations are a source of primary Particulate Matter (PM) and of ammonia, which is a main precursor for the forming of secondary aerosols in the atmosphere (table 1). Also on a local scale single intensive livestock installations or a high density of installations may contribute to a large extent to the PM immission load (Bleeker A. et al. 2007). So the control of PM emission is also in the focus of the licensing processes for the construction and operation of livestock installations under the Federal Immission Control Act (BImSchG).

2 Legal framework

In Germany, the legal basis for the protection of human health and environment against aerial pollutants is laid down in the Federal Immission Control Act (Bundes-Immissionsschutzgesetz – BImSchG 2002). According to the BImSchG livestock installation shall be constructed and operated in such a way, that this does not involve harmful effects on the environment or other hazards, considerable disadvantage and considerable nuisance to the general public and the neighbourhood (principle of protection).

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.

In detail, requirements are laid down in the First General Administrative Regulation Pertaining the Federal Immission Control Act (Technical Instructions on Air Quality Control – TA Luft of 24 July 2002). By this way several European Directives (esp. IPPC, NEC and European Air Quality Daughter Directive) have been adopted to German law. The requirements, especially emission and immission limit values and emission abatement techniques must be adhered to if industrial plants and other installations such as animal husbandries are constructed and operated, enlarged or in any other way substantial altered.

[1] Association for Technology and Structures in Agriculture (KTBL), Darmstadt, Germany

In order to verify whether a livestock building project complies with these requirements, data on the emission of particulate matter are needed. For the measurement of these emission is very laborious and costly, data derived from scientific investigations are used.

Table 1:
Emission of primary particles from agriculture and animal husbandry respectively

Type of emission	Total emission of Germany (2005) [kt/a]	Agriculture [kt/a]	Agriculture [%]	Animal husbandry [kt/a]	Animal husbandry [%]
Ammonia[1]	619	590	95	494	79
Total dust	269[1]	–[3]	–[3]	–[3]	–[3]
	513 (1995)[2]	71	14	49	10
PM10	194[1]	20.5	10	19.3	8
	281 (1995)[2]	23	8	22	8
PM2.5	111[1]	4.7	4.2	4.7	4.2

[1] UBA (2007)
[2] Klimont Z. et al. (2002)
[3] no data available

3 Available data on PM emission

Most of the data that are available and used for the assessment of environmental effects of livestock installations have been published for the inhalable fraction according to DIN EN 481 for occupational health and safety purposes. Those data are often used equal to total dust data although the precipitation for particles > 100 µm is only about 50 % in those measurements. Table 2 summarizes the results of different investigations in Europe showing a wide span.

Only few investigations have been carried out to determine PM10-concentrations. The proportion of PM10 is estimated pragmatically by so-called conversion factors that are derived from only few investigations (table 3).

Table 2:
Mean emission rates for inhalable dust (total dust), converted from the results of Takai H. et al. (1998) for Demark, Germany, England and the Netherlands; amended with data from Brehme G. (2003), LFL (2004) and Hinz T. (2005), summarized in KTBL (2006)

Animal category and housing system	Mean emission rate of investigations in Germany		Range of data of all investigations[1]	
	[mg/(AP h)][2]	[kg/(AP a)][2]	[mg/(AP h)][2]	[kg/(AP a)][2]
Pig fattening[3]				
- solid manure system	-	-	73 - 116	0.6 - 0.95
- liquid manure system	69	0.57	54 - 116	0.45 - 0.96
Sows[4]				
- solid manure system	226	1.9	43 - 226	0.36 - 1.9
- liquid manure system	49	0.4	36 - 255	0.31 - 2.14
Piglet rearing[5]				
- liquid manure system	22	0.18	21 - 41	0.17 - 0.34
Cows				
- solid manure system	91	0.8	72 - 170	0.6 - 1.5
- cubicle (liquid manure system)	406	3.6	25 - 406	0.2 - 3.6
Bull fattening				
- solid manure system	95	0.8	25 - 95	0.2 - 0.8
- liquid manure system	82	0.7	55 - 101	0.5 - 0.9
Calves				
- solid manure system	43	0.4	19 - 43	0.2 - 0.4
- liquid manure system	58	0.5	19 - 58	0.2 - 0.5
Laying hens				
- cage housing	2	0.02	1.4 - 3	0.01 - 0.03
- floor housing	40[6]	0.35[6]	6 - 14.8	0.05 - 0.13
- aviary (with aerated manure belt)	30[6]	0.27[6]	-	-
Broilers[7]				
- floor housing	4.2	0.03	2.8 - 9.3	0.02 - 0.06
Turkeys8)				
- floor housing	73	0.55	-	-
Ducks				
- rearing[9]	1.7	0.012	-	-
- fattening[9]	5.4	0.04	-	-

[1] Denmark, England, the Netherlands and Germany
[2] AP = animal place
[3] 343 days housing period
[4] 350 days housing period
[5] 346 days housing period
[6] LfL (2004)
[7] 264 days housing period
[8] turkey cock, 314 days housing period; Hinz T. (2005)
[9] 300 days housing period; Brehme G. (2003)

Table 3:
Proportion of PM10 of total dust (KTBL 2006)

Animal category and housing system	PM10-ratio of total dust		Source
	Measured data	Convention	
Pigs	0.31 0.45 0.4	0.4	Koch W. et al. (2002) Louhelainen K. et al. (1997) Berdowski J. J. M. et al. (1997)
Cattle	0.3 0.46 0.64 (0.33-0.87)	0.5	Koch W. et al. (2002) Seedorf J. (2003) Cathomas R. L. et al. (2002)
Poultry (in general)	0.46 0.4 0.15-0.16		Koch W. et al. (2002) Berdowski J. J. M. et al. (1997) Cravens et al. (1981)
Laying hens (cage housing)	0.33	0.3	TÜV Süddeutschland (2000)
Laying hens (aviary housing)	0.5 0.6	0.5	Hinz T. (2005) LfL (2004)
Laying hens (floor housing)	0.59	0.6	LfL (2004)
Broilers	0.4 0.58	0.5	Hinz T. (2005) TÜV Süddeutschland (2000)
Turkeys	0.25	0.3	Hinz T. (2005)

3 Provisions on air pollution control

The TA Luft (2002) determines limit values for the immission of fine particles (PM10), which are cited from the European Air Quality Daughter Directive (European Union 1999) (table 4).

Table 4:
Immission limit values of TA Luft (2002) for PM10

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]
40	1 year	-	< 34	1.2
50	24 hours	≤ 35 exceedances per year permissible	≤ 15 exceedances per year over the last 3 years	-

During the licensing process of a livestock installation project the assessment whether those limit values are not exceeded is necessary only for those places of interest, where within a distance of 1 km from the installation people could not only temporarily be exposed to PM immissions, as it is the case e.g. in dwelling areas.

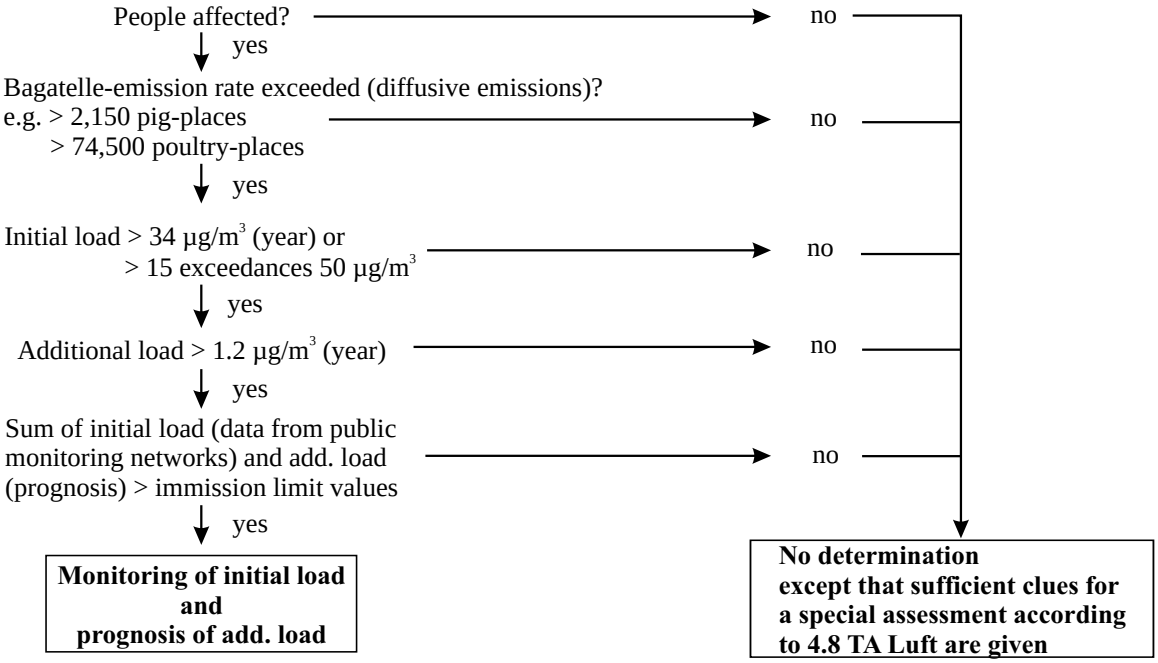

Figure 1:
Assessment scheme relating to PM10 immission according to TA Luft (2002)

For reasons of commensurability the competent authority shall dispense with the need to determine the immission load in detail, if in the case

- of a low emission mass flow rate (cf. 4.6.1.1 TA Luft) or
- of a low initial load (cf. 4.6.2.1 TA Luft) or
- of an irrelevant additional load caused by the installation (cf. 4.2.2 TA Luft) and additional measures for emission abatement

it can be assumed that harmful effects on the environment cannot be caused by the installation. The examination follows the scheme illustrated in figure 1.

In contrast to the regulations to control the immission load, regulations of the TA Luft on emission limitation for total dust in general will not affect livestock installations.

3.1 Low emission mass flow rate

Immission of PM10 are usually not determined, if the emission of a livestock installation do not exceed the so-called bagatelle-emission rate of 0.1 kg/h of total dust. The bagatelle-emission rate is equivalent to a livestock of about e.g. 2,150 fattening pigs and 74,500 laying hens in conventional housing systems taking the rounding adjustment (0.149 kg/h, cf. 2.9 TA Luft) into account.

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.

3.2 Low initial immission load

The existing initial immission load of PM10 is low, if the annual mean does not exceed 34 µg/m³ and if the daily immission load of 50 µg/m³ is not exceeded for more than 15 days a year over the last three years.

Table 5 gives an overview on the concentration of PM10 in different regions of Germany. Highest levels are usually monitored in congested urban areas and at sites influenced by and close to major roads.

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. This is also indicated by the distribution of measured PM10 concentrations, which show a strong correlation between the annual mean value and the number of days exceeding 50 µg/m³ (figure 2).

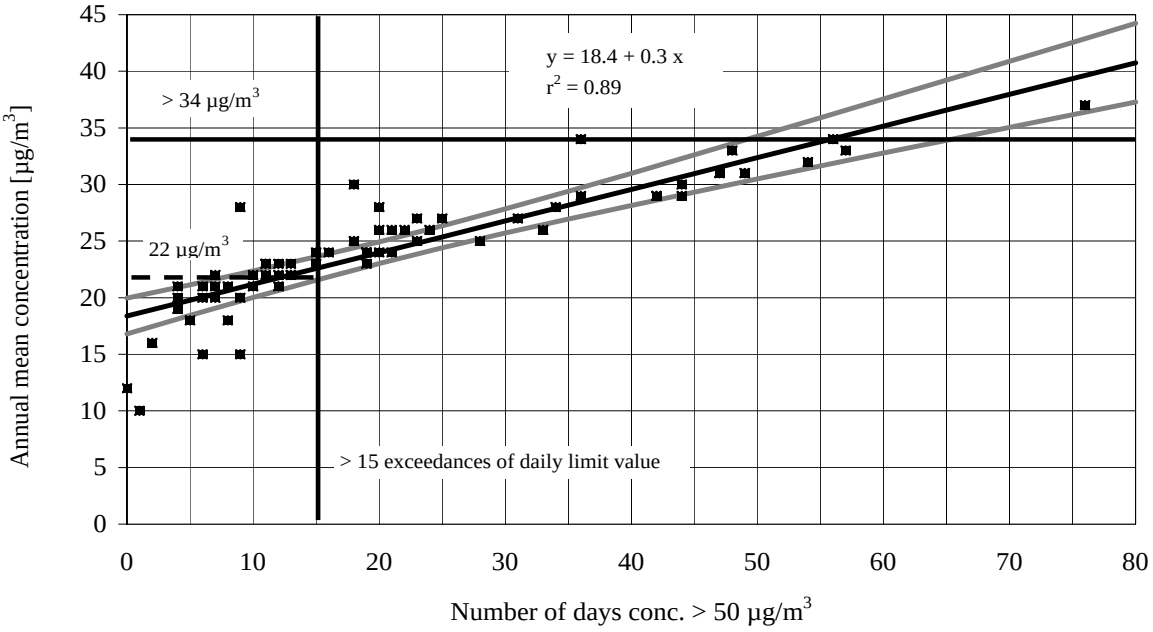

Figure 2:
Correlation between the number of days exceeding the daily limit value of 50 µg/m³ and the annual mean concentration of PM10; regression line and confidence interval for the mean value with 99 % confidence level (Lower Saxony 2003 – 2005; Niedersächsisches Umweltministerium 2006)

Table 5:
Typical ranges of concentrations of PM10 at different locations in Germany 2001 (Lahl U. 2005)

Location	Rural site	Urban site	Traffic site
Annual mean concentration [µg/m³]	10 - 18	20 - 30	30 - 45
Number of days exceeding the daily limit value of 50 µg/m³	0 - 5	5 - 20	15 - 100

As the level of the mean annual concentration for PM10 is more homogeneous in the wide-range and easier to prognose than the number of days exceeding the daily limit (Umweltbundesamt Wien 2005), with the help of this graph annual mean concentrations can be transposed to a number of exceedances of the daily limit value (Fierens F. et al. 2006; Torfs R. et al. 2007). In the case of Lower Saxony it can be stated with a 99 % confidence level, that the number of exceedances of the daily limit value will be below 15 if the annual mean concentration measured or prognosed in rural areas is below 22 µg/m³.

3.3 Irrelevant additional immission load

Despite high initial loads a project might be licensable if the additional load caused by an installation is classified as irrelevant (1.2 µg/m³) in terms of TA Luft (2002). This level 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/m³ (Seedorf J. and Hartung J. 2002), must be diluted by a factor of 1,000.

The modelling of the dispersion of fine particles as prescribed in the TA Luft (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 (figure 3). It can be stated, that e.g. in the case of pigs the irrelevant distance for PM10 is smaller than for ammonia and even smaller than for odour according to the minimum distance regulation of the TA Luft (2002) to prevent odour compliances (cf. 5.4.7.1 TA Luft).

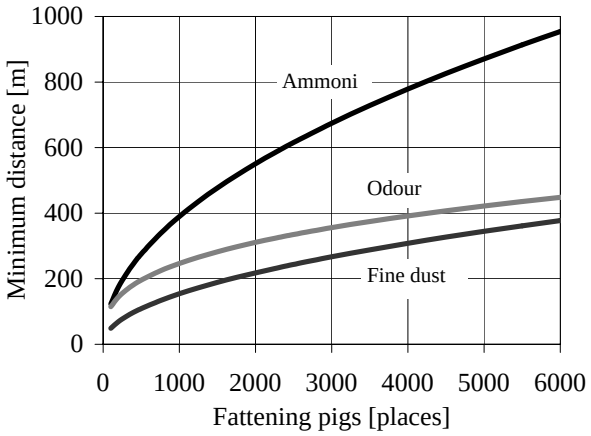

Figure 3:
Minimum distances for ammonia and odour according to TA Luft for fattening pigs. The irrelevant distance for PM10 (1.2 µg/m³ additional load) is calculated on the same assumptions as for ammonia.

4 Conclusions

In Germany emission of PM are controlled during the licensing process of livestock installations. In practise only the regulations of the TA Luft concerning the limitation of immission of PM10 are relevant, for the emission limit values, that are applying to total dust, are not exceeded in livestock installations.

There are some provisions in order to simplify the execution of the immission load regulation, but they do not take effect in all cases. For example, the bagatelle-emission rate that equals to installations with 2,150 pigs or 74,500 laying hens in conventional housing systems, in the case of alternative housing techniques such as littered pens or floor housing systems may be exceeded by even a smaller livestock. In addition, the criteria of the TA Luft 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, if a livestock installation e.g. in the case of an enlargement or any other substantial alteration does not keep the minimum distance regulations towards the next dwelling area, additional measures for emission abatement must be taken.

For the adequate prognosis of the emission and the immission caused by a livestock installation reliable and differentiated emission data are missing. Up to now measurements are 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 and/or the respirable fraction. Hardly any data have been published for PM10 emission. 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. There is an urgent need to establish and harmonise sampling procedures for the measuring of emission of particulate matter from livestock housing systems.

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PM emissions and mitigation options from agricultural processes in Germany

O. Beletskaya[1], I. Huck[2], E. Angenendt[1], J. Theloke[2], J. Zeddies[1], and R. Friedrich[2]

Abstract

Agriculture is one of the sources of particulate matter (PM) emissions to the atmosphere. For calculating emissions and assessing abatement potentials of mitigation measures, an economic-ecological modeling system is elaborated. The main target of the model is the improvement of the German agricultural inventory. Primary PM with fractions of 10 and 2.5 µm is taken into account, because of a high concern for the environmental policy due to relevant health impacts caused.

Abatement strategies for the particulate matter emission will be formulated and assessed in the framework of the regional model, **EFEM** (**E**conomic **F**arm **E**mission **M**odel). Five regions in Germany will be considered: Brandenburg, Niedersachsen, Bayern, Nordrhein-Westfalen and Baden-Württemberg.

In order to get results on the PM emission for these regions, activities (animal husbandry, crop production, fodder preparation, manure management, tillage, land preparation and harvesting operations) of different farm of various types and information on the emission intensities will be compiled with the help of linear programming. Emission factors will be either determined or taken from studies about European agricultural particulate emission and adopted for the **EFEM**. The emission outputs of the model will be implemented into the IER emission model, which is able to calculate high spatially resolved emission data.

The effects of mitigation options to the PM emission from agricultural activities in Germany will be assessed on the base of comparison of the reference year 2003 and scenario 2013.

Finally, implementation strategies of the most efficient mitigation options will be elaborated, regarding technical and non-technical measures in particular under consideration of the economic impacts to the recipients and other regional effects.

Keywords: particulate matter emission, economic-ecological modeling system, abatement strategies

1 Introduction

The contribution of agriculture to the total particulate matter (PM) emission in 2004 - 2005 in Germany was about 9 per cent of the PM10 and nearly 7 per cent of PM2.5 primary emission (Umweltbundesamt). PM with particle sizes of 10 and 2.5 µm is of the highest interest for both political and healthcare organizations. On one hand, information on the emission of the above-mentioned PM fractions is more desirable, because already existing data about it are not enough to fulfill the international reporting obligations, to monitor progress in air pollution and to identify possible mitigation strategies. On the other hand, many studies highlight, that particles of 10 and 2.5 µm, associated with inhalable dust (in some researches – with inhalable and respirable dust respectively), cause serious health problems. In this relation data on PM10/PM2.5-emissions are quite essential for further development of technologies (e.g. BAT - **B**est **A**vailable **T**echnologies) effectively abating PM emission from agricultural systems, as well as of political measures directing agricultural management to a more sustainable way.

2 Objectives

The main objective of the project “Modeling of sectoral, spatial disaggregated balances of greenhouse gases and assessment of environmental protection strategies at the regional policy level” financed by **German Research Association (DFG)**, is to develop an improved German agricultural inventory for ammonia and greenhouse gases emissions. PM-emission, with respect to its assessment, development of possible abatement measures and determination of their effects on other environmental indicators, has become a key issue of this study.

As the result of the research following sub-targets are to be attained:

- Representation of emission “hot-spots” in regions and consequent choice of experiment regions.
- Modeling of the primary PM emission on the level of selected regions and their administrative districts under the consideration of various local conditions and farm structures.

[1] University of Hohenheim, Institute for Farm Management, Germany

[2] Universität Stuttgart, Institute of Energy Economics and the Rational Use of Energy, Germany

- Spatial and temporal distribution of PM emissions in experiment regions
- Appraisal of future emission development and evaluation of abatement possibilities (their potentials, costs, other disadvantages and advantages) and therefore their ordering according to efficiency and political feasibility.
- Transfer of the results for chosen regions on the perspective of the whole of Germany.

In order to reach these objectives an economic-ecological modeling system will be elaborated, also extended, improved, and finally implemented.

3 Research regions

Five regions are taken into consideration: Niedersachsen, Brandenburg, Bayern, Baden-Württemberg and Nordrhein-Westfalen. Each county is subdivided into administrative districts (Regierungsbezirke).

The above-mentioned regions have been chosen for the study, because "hot spots" of emission can be pointed out according to the emission registry, and the choice of the mentioned German counties was made with regard to these "hot spots". To demonstrate the importance of various agricultural emission sources for primary PM, it does make sense to choose regions with different key production activities. Thus, Niedersachsen and Nordrhein-Westfalen are considered because of their intensive livestock production. According to the report of the German Environmental Organization, the highest amount of PM is emitted from livestock farming; in 1995 the emission from animal husbandry in Europe amounted to 4.5 per cent of PM10 and 1.7 per cent of PM2.5 (Umweltbundesamt). Other most important emission sources are fodder, dry manure (uppermost in the poultry houses) and litter. Bayern and Baden-Württemberg were chosen, because of high shares of forage-growing activities presented there (50 and 36 per cent of forage-growing farms accordingly). Besides animal production, PM emissions are registered as well from other kind of agricultural activities, for instance, from arable farming. In this relation Brandenburg was regarded as the region of extensive arable crop production with a high share of arable farms (34 per cent), carrying out their agricultural activities on 38 per cent of the total used agricultural land. It is also important to mention that in all selected regions from 40 to 55 per cent of land is under agricultural use. So, the elaboration of PM-emission mitigation policy looks quite essential for these German counties, concerning the information presented above (Destatis).

4 Methods and materials

4.1 Economic Farm Emission Model (EFEM)

Measurement of PM-emissions at the farm scale is feasible and models allow the most coherent estimating of emissions at the regional scale. However, in many models measuring of PM-emission is associated with great technical requirements and high costs. Recently, PM-emissions caused by agricultural activities are considered in multi-sectoral models (e.g. **RAINS - R**egional **A**ir Pollution **In**formation and **S**imulation). Anyway, by the reason of their complexity, detailed examination of the agricultural sector is hardly possible. That is why an ecologic-economical **E**conomic **F**arm **E**mission **M**odel (**EFEM**) was developed (Schäfer M. 2006).

EFEM was used for various projects and, therefore, its key research issue has been continuously extended. Thus, the current work is dedicated to analyze PM-emission from various agricultural systems.

In the model, the target of the economic analysis carried out on the farm-level corresponds to the farmer's interests. Therefore the objective of **EFEM** is the maximization of an individual farm's gross-margin. These farms are regarded as representative for all farms of the same type in the same region, and their objective function can be written as follows:

$$\max_{\boldsymbol{x}_k} \boldsymbol{\pi}_k = (\boldsymbol{p}_i - \boldsymbol{c}_i) * \boldsymbol{x}_k,$$
$$\text{s.t. } \boldsymbol{A}_k * \boldsymbol{x}_k \leq \boldsymbol{z}_k, \boldsymbol{x}_k \geq 0,$$

where $\boldsymbol{\pi}_k$ is the total gross-margin of the k-th farm-type, $\boldsymbol{p}_i$ and $\boldsymbol{c}_i$ are n-vectors for the i-th selling price and variable costs of the i-th production activity consequently, $\boldsymbol{x}_k$ is the n-vector of command variables. The constraints faced by farm-type k are defined through $\boldsymbol{A}_k$ and $\boldsymbol{z}_k$, determining $m \times n$-input-output matrix and m-vector of capacities accordingly.

Command variables in **EFEM** are area associated to each crop (including grassland and set-aside area), the number of animals from different categories, and quantity of purchased animal feeding, variables related to policy programs (enrolment in environmental or set-aside programs), total greenhouse gases emissions. In the letter case $\boldsymbol{p}_i$ and $\boldsymbol{c}_i$ are substituted by the tax on emissions (De Cara S. et al. 2006).

EFEM is a mixed integer model, one of special cases of linear programming, where the corresponding variables can be included as both binary and integer. The given approach reflects the possibility of farmers to deal in mutually exclusive policy programs including different obligations and payments. **EFEM** takes into account crop and grassland

products and crop by-products, which can be used on farm for feeding purposes, for litter, or green manure selling (De Cara S. et al. 2006).

There are the following constraints defined by A_k:

1) Cropping area
2) Crop rotation
3) Animal housing places and animal demography
4) Livestock feed requirements
5) Quotas
6) Restrictions imposed by policy and environmental programs

In **EFEM** total land is fixed at initial land endowment of each farm-type. Crop rotation constraints relate to maximum area shares of certain crops in a rotation process. Based on historical observations and calibrated to reflect common practices and rationality of agricultural business, these constraints aim at substituting the dynamics of crop rotation and thereafter translating it into a static model. Livestock numbers are also limited by the availability of fixed capital (number of housing places). As for modeling of animal feed requirements, the minimum requirements of energy and protein for each livestock category are to be met, as well as for digestible matter. Among quotas sugar beet and milk quotas are distinguished as constraints. Set-aside requirements fell into the category of restrictions imposed by policy end environmental programs (De Cara S. et al. 2006).

The core of **EFEM**, showing an interaction of both economic and environmental elements, is the production module. The model is adapted to find simultaneous solutions of multidimensional problems and adjusted for various farm-structures. Last-mentioned were chosen according to the business alignment (**BWA** – **B**etriebs**w**irtschaftliche **A**usrichtung), with the only exception: horticultural business was excluded from the modeling. Agricultural activities of the following 5 farm-types will be regarded: arable farming, permanent cropping, forage-growing, intensive livestock, and mixed farms.

The model is composed of different sub-models, making possible a representation of all relevant production processes of arable farming and grassland management, with related mechanization, animal husbandry with a disaggregated feeding module, manure management and nitrogen circulation. Besides these mentioned constituents, production modules of each mentioned sub-module also contain energy, emission and stock flow parameters for quantification of emissions caused by the above-mentioned operations. Moreover, greenhouse gases and PM emissions are differentiated according to the production branch, which these gases and PM outflow from (see figure 1).

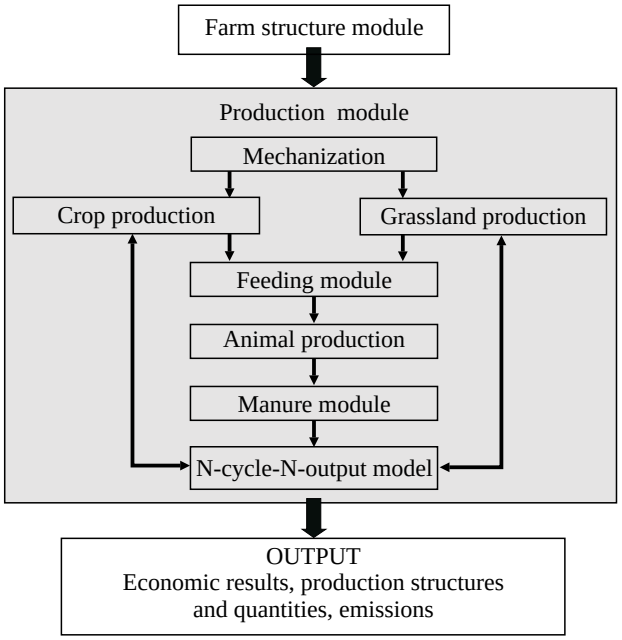

Figure 1:
Build-up of the EFEM (Triebe S. 2007)

Costs and activities data such as yield, productivities, and intensities are different from region to region. All production activities presented in **EFEM** are highly disaggregated according to the level of input-use and performance. Firstly, in general 14 different arable crops are integrated into **EFEM**: winter- and summer-wheat, winter- and summer-barley, oats, rye, winter-rape, sugar beets, potatoes, field beans, sunflowers, grain maize and silage corn, as well as clover-grass. Each crop-production activity can be combined with different fertilization intensities. Secondly, **EFEM** distinguishes between two grassland activities: meadow and pasture (Triebe S. 2007; De Cara S. et al. 2006). Thirdly, livestock production is also subdivided according to levels of performance for main animal categories kept in selected regions. The part of the model devoted to animal husbandry includes data on dairy farming, calves, heifers, bulls, fattening pigs, cows, sheep, and intensive livestock farming with emphasis on hog and poultry production (Triebe S. 2007).

PM-emissions are differentiated in **EFEM** for following agricultural activities: land preparation, seeding, harvesting, livestock as well as forage production and manure management. Therefore, emission factors for these production activities and the operations they comprise are considered in the model. For instance, emission intensities for such operations of arable farming as land preparation, crop production, and harvesting are accounted for. In the case of feeding and manure managements the production of feed and manure consequently is considered. For animal hus-

bandry various types of manure and housing systems are taken into account while determining emission factors.
Information on the emission intensity and farm activities data, integrated into **EFEM** allows analyzing and predicting the development of PM emission firstly on the farm level. With the next step of modeling due to numerous model-calculations, technical and political abatement strategies with the accent on their reduction potentials are examined on the same level. Targeted technical and political options are to be oriented to assess PM emission and, at the result, to find out a proper ways to minimize or even eliminate it.

In **EFEM** optimized results for individual farms will be extended to the regional level due to a special extrapolation procedure.

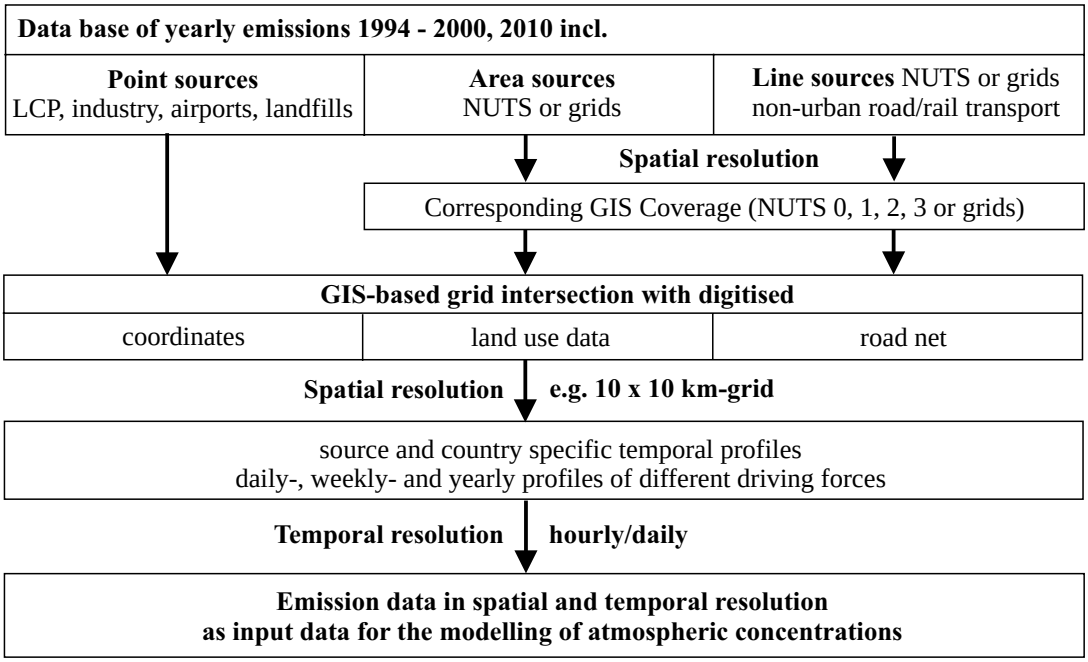

Figure 2:
General approach for the generation of emission data in high spatial and temporal resolution within the **IER** emission model (Pregger T. et al., 2007)

4.2 IER ECM-model

The information on PM-emission resulting from the **EFEM**-model will be implemented into the **IER emission calculation model (ECM).** The latter is developed to calculate emission data with a high spatial resolution. The basic methodology of the **IER** approach is shown in figure 2.

The model starts with a yearly emission table that includes point, area and line sources using different geographic data for spatial allocation of these source types. Spatial allocation based on GIS intersection leads to grid- and cell-shares of emission sources considered. PM emissions in areas selected for the research will be calculated in a spatial resolution of 5 km x 5 km. Results will be parameterised, so that the emissions in other regions of Germany can be assessed. The spatial resolution of the GIS regarding the whole of Germany will be 10 km x 10 km. If more than one county is located in a grid segment, the respective emissions of each county will be weighted by the relative size of its area, resulting in the average emissions of the segment. The resulting shares are stored in two database tables and used by the **ECM** for multiplication with the yearly emission data and temporal profiles.

5 Source data

To build up the regional typical farm-model, bookkeeping data for individual farms in the considered German counties for the reference year 2003 are needed. This information is obtained from **F**arm **A**ccountancy **D**ata **N**etwork (**FADN**). Test-farms are to be assessed on the regional level and, thereafter, averaged. For each region up to 5 typical farms, whose factor endowments are close to average results, will be selected.

FADN databank contains consistent information on costs, prices, areas, and income for all selected regions and administrative districts of Germany. **FADN** provides infor-

mation on important agricultural products (meat, milk, crops, etc.), but does not take into account any changes of production intensities of a single production process. For each farm type input and production intensities are fixed throughout prices, costs and yields for a certain farm-type. In return that means that the use of **FADN** data requires an analysis of the changes of agricultural management induced by deviations from the original one. This divergence has significant influence on fertilizer application, cultivated crops and so forth (Vabitsch A. M., 2006).

At the result of a comparison between data from **FADN** and regional statistics, significant differences were found. Moreover, figures from **FADN** for the same parameters and regions are lower than those from the regional statistics. Such a difference can be explained by the fact that the bookkeeping information for part-time farms is not taken into account in the **FADN**-database. To be able to project the results from farm to regional level, a methodology for extrapolation was developed. It is based on an optimization approach, which eases the representation of total capacities of regarded regions through the weighting of capacities of typical farm-types. In **FADN** alternative extrapolation factors expressing number of representative farms are used. But for the case of **EFEM** these farms cannot be used as extrapolation coefficients, firstly, because of the deviation from regional statistics. Therefore extrapolation factors are being elaborated according to the approach presented by Schäfer M. (2006). Its basic principle presumes the presentation of a serial of typical model farms, their assessment and weighting in order to match them to the existing regional production and farm structures and, therefore, to represent regional capacities exactly. Rather typical than average farms will be extrapolated in this study. Each of the regional capacities will be weighted with the respective gross-margin to guarantee that they won't be overestimated. Monetary deviations, i.e. sums of absolute under- and overestimations are to be minimized through the optimization approach (Schäfer M. 2006).

Factor endowments for each region will be exactly assessed due to the multiplication of the adjusted farms capacities with the weighting coefficient. Extrapolation procedure additionally assures that farm structures in the selected counties are well represented.

On the basis of bookkeeping data together with information on PM-emission intensities, the presentation of a German detailed agricultural inventory for PM10/PM2.5-emissions from agriculture is to be done. It is the main concern of the work in the described research project. Emission-factors will be provided by **IER** on the base of own calculations and literature research.

The choice of PM emission factors has appeared to be a quite problematic issue, mostly because of the absence of a worldwide unique inventory system. Information on emission intensity for different PM fractions, measurement aggregations and units make it difficult to use emission data for the final modeling without additional preprocessing.

There is still not enough information to conclude about the contribution of, for example, arable farming with relevant operations of land preparation, crop production and harvesting. Even if there is any information on PM-emission intensity, very few studies analyze how uncertain emission factors can be calculated and, therefore, comparison of information on emission from different researchers shows not always clearly where emission data are overestimated and where they are underestimated.

Very often European emission is assumed as being equal or at least equivalent to the American one without consideration of different climate and soil conditions. The uppermost reason is that the emission factors are more readily available in American studies than in European ones. This holds particularly true for emission studies from California. Due to the lack of relevant European research the contribution of some agricultural activities to the total PM emission is still not known as, for instance, in the case of operations on land preparation.

However emission data from some sources are already used in big projects and can be used as well to build up the current model on the PM-emission. Thus, in the **RAINS**-model developed at the **I**nternational **I**nstitute for **A**pplied **S**ystems **A**nalysis **(IIASA)** the existing structure for assessment of gaseous emissions was already extended to estimate PM-emission. For an appraisal of the PM-emission from animal production and arable crop production the most important animal categories and arable farming activities have been chosen, in other words, the aggregation of parameters, regarded as PM-emission sources, in **RAINS** is very general. The elaboration of eventual emission factors in this model is based on the results of two studies: Takai H. et al. (1998) and ICC&SRI (2000).

Some studies from the German Federal Agricultural Research Centre (**FAL**) and the Leibniz-Centre for Agricultural Landscape and Land Use Research (**ZALF**) in turn provide sufficient information on emission from the arable farming. Here the aggregation of PM-emission sources according to different operations is relatively detailed.

The study of Seedorf J. (2004) is another source of information on emission from animal production. Measuring techniques used for this work are different from those applied by Takai H., et al. (1998). This is possibly the most significant reason for differences in the two studies' results. Anyway, emission is measured for inhalable and respirable dust in both works. But if in RAINS inhalable and respirable dust from Takai H., et al. (1998) was associated with PM10 and PM2.5 accordingly, Seedorf J. (2004) suggests transformation factors in order to obtain PM10 and PM2.5 emphasizing that these conversion data should be used very

carefully, because there is no clear correlation between inhalable and respirable dust and the above-mentioned PM fractions. Moreover, Seedorf J. (2004) suggested to take into account housing periods to make data more precise and presented the way how these data can be calculated. Klimont Z. and Amann M. (2002) (IIASA) have also suggested to elaborate emission factors considering housing period but the way of its calculation is not clear.

6 Expected results

The most prominent strength, responsible for the results expected from the implementation of the **EFEM**-model, is the possibility to calculate simultaneously both consequences of abatement strategies and development of PM emissions.

The model allows an appraisal of emissions from agricultural systems on farm and regional levels. Calculated results for the PM-emission will be differentiated according to the farm-type and regional conditions. At the result of a sophisticated regional analysis, strategies with the most efficient mitigation options will be chosen, regarding technical and non-technical measures, for the simulation of how agricultural policies (for instance subsidies, law) and socioeconomic framework (prices, labour, etc.) influence farmers' decisions on management options and on intensity of agricultural activities, that eventually affect the expected revenue. Thus, these abatement strategies are to be evaluated through measurements influencing either activities or specific emissions of each activity unit (emission factors), with respect to environmental effectiveness and economic feasibility.

Due to the whole modeling process, depiction of spatial distribution of PM-emission "hot-spots" will be represented through the implementation of GIS. Regional results are to be extrapolated for the whole of Germany. Therefore, the development of an improved German emission inventory is expected from the implementation of the **EFEM**-model. An emission inventory for PM will be presented for the year 2003, effects of mitigation options in Germany will be appraised on the base of the reference scenario for 2013, comparing calculated emission with emissions in a reference scenario. With the scenario 2013, future PM-emission from agriculture will be evaluated under consideration of agricultural policy development.

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Agricultural particulate matter emissions in the Czech Republic

H. Hnilicova[1] and P. Hnilica[2]

Abstract

Recent studies have demonstrated adverse effect of particulate matter on human health. Area of the Czech Republic, where is annual limit value of PM10 exceeded, permanently increases. In order to prepare effective measure of air quality improvement we have to compose complete emission inventory. One item, that in Czech emission inventories is missing, is emissions of agriculture activities. On preparation emission inventories are national emission factors preferred. In the Czech Republic no agriculture emissions were realized, therefore we will adapt USEPA method.

Basic operations on arable farming are:

- soil preparation
- harvesting
- transport
- unloading
- post harvest treatment

Emissions from transport, unloading and post harvest treatment are included in other source categories for that reason we will deal with soil preparation and harvesting. According to USEPA emission factors from this operations are depended on

- soil type respective its silt content
- weather conditions
- species of vegetal products
- method of soil preparation

All the above items will be incorporated into emission factors.

For emission estimation from animal farming it was taken into account species and age structure of animals in the Czech breeds and breeding technique.

Keywords: asrable farming, animal farming, PM emissions, agricultural operations

Introduction

Agriculture particles are emitted from both animal and plant production. Their composition is very various and depends on particle origin. Particles from plant production include soil minerals, organic material from plants, pollen and fungal spores. From animal production there are particles coming from feed, rest of skin and hairs and also dry manure (Atmospheric emissions of particulates from agriculture: a scoping study, 2000). Agricultural bioaerosols cause respiratory disease and some matter presented in this aerosol cause allergic reaction of skin, eye and nasal mucosa. Agricultural emissions come from housed livestock, arable farming, crop storage, energy used on farms and unpaved roads on farm. The emission from energy used and crop storage have been included in our inventory for that reason we try to estimate emission from housed livestock and arable farming in this paper. The emission estimation from housed livestock is based on methodology presented in AEI Guidebook (2006). Since the chapter on arable farming emissions is not available the emission estimation for this source category is based on USEPA methodology.

Arable farming

The current version of AP-42 (i.e., the 5th edition) does not address agricultural tilling even though a PM10 emission factor for fugitive dust generated by agricultural tilling was developed by Midwest Research Institute in 1983 and adopted by the USEPA in their 4th edition of AP-42. Thus, the methodology adopted by the California Air Resources Board (CARB) (Countess Environmental (2004) WRAP Fugitive Dust Handbook) is presented as the primary emissions estimation methodology in lieu of an official EPA methodology for this fugitive dust source category.

The way of tilling fields, the soil and climatic conditions in Mid-West is very different from conditions in the Czech Republic therefore it was necessary to adapt this emission model.

The USEPA (NATIONAL AIR POLLUTANT EMISSION TRENDS, 1900 – 1998, 2000) has published a general empirical formula for dust emissions due to land preparation, which can be used to estimate the amount of particulate matter (PM10, PM2.5) generated per acre-pass.

[1] Czech Hydrometerological Institute, Prague 4, Czech Republic

[2] Private researcher

$$E = k \times S^{0.6} \times a \times c \times p \qquad 1$$

where

S = % silt content of the soil defined as the mass fraction of particles smaller than 75 μm diameter found in soil to a depth of 10 cm (%)

a = field area

c = constant emission factor (c = 4.8 lbs/acre-pass)

p = number of tilling

The amount of emission depends on soil type only not on the type of working operation for this type of emission estimation. Modified model takes the type of working operation into account but there is no dependence on character of soil under the plough (cultivated land). For this reason the following relation for emission estimation is applied.

$$E = k \times S^{0.6} \times \sum a_j \times \left(\sum EF_i\right) \qquad 2$$

where

E = PM emissions

k = dimensionless particle size multiplier (EPA default is for PM10 = 0.21, PM2.5 = 0.042)

S = weighted mean % silt content of the soil in the Czech Rep.

a_j = areas under farm crops for each individual crops

$\sum EF_i$ = sum of emission factors for individual working operation using during the field works performed for the most frequently grown plants during the year

Nature conditions of Czech Republic

The Czech Republic is a landlocked country located in moderate geographical latitudes in the Northern Hemisphere. The climate of the Czech Republic can be labeled as moderate, of course with great local diversity seen throughout the year. The most important factor in the diversity of the Czech climate remains the varied topography, thanks to which the climate varies among individual regions of the country. These factors cause regional differences of air temperature, precipitation amount, winds and consequently the variability of soil moisture.

Soil types and methods of their agriculture cultivation

All above mentioned climatic factors influence the methods of agriculture soil cultivation. The way of soil preparation depends on the amount of precipitations in autumn. This is the time for sowing of winter wheat and rape. Generally speaking the dryer autumn the more procedures are needed for land preparation for sowing.

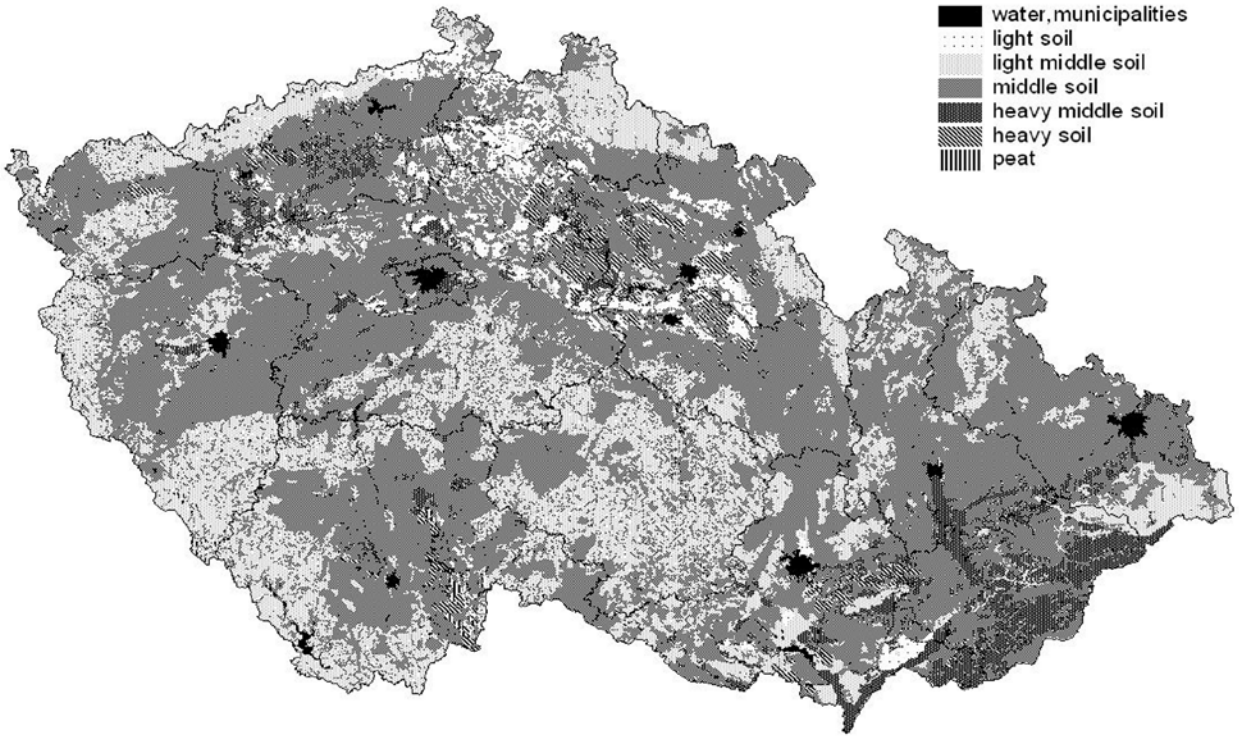

Figure 1:
Soil map of Czech Republic

Another factor that influences agriculture soil cultivation and PM emission during the field works is the soil type. The soil map of the Czech Republic is presented on figure 1.

According to the Czech classification (Petr J., et al. 1989), the major part of soils in the Czech Republic falls in category "middle". This category implies "loam" and "silt loam" categories according to triangle classification. The Czech soil classification and the triangle one are compared in table 1. The weighted mean of silt content amounts to 40 % in the Czech Republic.

Table 1:
Soil classification

Czech Soil Type	Soil Type texture triangle	Silt Content (%)
light	sand	12
light middle	loamy sand	12
	sandy loam	33
middle	silt loam	52
	loam	40
heavy middle	clay loam	29
heavy	clay	29

On the base of data from the Statistical yearbook (2006) there were selected crops whose areas under crops cover about 80 % of arable soil in the Czech Republic. For these corps there were specified the typical operations of land preparation. These operations were chosen from Czech publications (Petr J. et al. 1989, Autorský kolektiv 1998, Pulkrábek J. 2003) and consulted with farmers (see table 2 and table 3). Emission factors were selected from the work (Bogman P. et al. (2007). Emission factors published in this study stem from measurements which were provided for the soil type with the silt content amounting to 40 %.

This value corresponds to the weighted mean content of silt in soils in Czech Republic. The equation (2) transforms into

$$E = k \times \sum a_j \times \left(\sum EF_i \right) \qquad 3$$

because formula $\sum EF_i$ includes of soil texture influence.

A soil preparation for seeding is divided into operations for spring and winter crops. The first ones always emit less dust particles then autumnal operations, because the soil is mostly moist by spring when they are performed. This was also the reason why the emission factor for floating has been decreased for spring compared to the autumn season. Furthermore, it should be mentioned that demands for working operations for particular crop may differ even within one farm and depend on factual soil conditions. Emissions for the Czech Republic were calculated according to formula 2, where we take B = 1 for S = 40 % as a typical value in the Czech Republic.

Table 2:
Agriculture operations for winter cereals and winter rape

Winter crops			
Wet autumn		Dry autumn	
Operation	TSP emissions (kg/ha)	Operation	TSP emissions (kg/ha)
root cutting	1.26	root cutting	1.26
discing, tilling, chiseling	4.5	discing, tilling, chiseling	4.5
fertilizing	0.6	fertilizing	0.6
floating	4.5	2x floating	9
seeding	4.5	seeding	4.5
spraying	0.6	spraying	0.6
harvesting	5.5	harvesting	5.5

Table 3:
Agriculture operations for spring cereals, corn and sugar beet

Spring crops	
Operation	TSP emissions (kg/ha)
root cutting	1.26
ploughing	4.5
fertilizing	0.6
floating + seeding	2.5
spraying	0.6
harvesting[1]	5.5
for corn and sugar beet cultivation	3

[1]without sugar beet

Results of arable farming emissions assessment for the Czech Republic

Results of assessment of emissions from arable farming in the Czech Republic are summarized in table 4 and compared with those estimated according to RAINS methodology (http://www.iiasa.ac.at/web-apps/apd/RainsWeb/RainsServlet1). The RAINS estimates show significantly lower values. The reason for this fact is a need for application of more operations, because only about one half of Czech farms are equipped with advanced technologies enabling matching of several working operations. Zero-tillage approaches are not appropriate for all regions of Czech Republic, especially for spring crops. These technologies are applied in limited extent because their application for heavy-textured soils is too energy-consuming.

Table 4:
Czech emission from arable farming, 2005

	Unit	Emissions	Emissions according to IIASA
TSP		55535	4990
PM10	Mg/year	11662	274
PM2.5		2332	0

The emissions from arable farming are not insignificant and furthermore they are concentrated on several months in year. Monthly variability in agricultural operations is showed in table 5.

Table 5:
Monthly variabilities in agricultural operations

Crop	Jan	Feb	Mar	April	May	Juny	July	Aug	Sept	Oct	Nov	Dec
winter wheat												
spring wheat												
winter barley												
spring barley												
Triticale												
Oats												
Rye												
Rape												
Grain maize												
Industrial sugar.beet												
Green and silage maize												
Other annual fodder crops												
Lucerne												
Red clover												
Other perennial fodder crops												

Table 6:
Emission from animal breading, 2005

		Ratio	EF PM10 kg/animal	EF PM2,5 kg/animal	Number 1000 head	PM10 Mg	PM2,5 Mg
Dairy cattle	Tied or litter	0.90	0.36	0.23	564.00	183	117
	Cubicles (slurry)	0.10	0.70	0.45	564.00	39	25
Beef cattle	Solid	0.90	0.24	0.16	598.00	129	86
	Slurry	0.10	0.32	0.21	598.00	19	13
Calves	Solid	0.90	0.16	0.10	212.00	31	19
	Slurry	0.10	0.15	0.10	212.00	3	2
Sows	Solid	0.00	0.58	0.09	229.00	0	0
	Slurry	1.00	0.45	0.07	229.00	103	17
Weaners	Solid	0.00	na	na	975.00	0	
	Slurry	1.00	0.18	0.03	975.00	176	28
Fattening pigs	Solid	0.00	0.50	0.08	1630.00	0	0
	Slurry	1.00	0.42	0.07	1630.00	685	112
Horses	Solid	1.00	0.18	0.12	23.00	4	3
Laying hens	Cages	0.95	0.02	0.00	6316.00	102	13
	Perchery	0.05	0.08	0.02	6316.00	27	5
Broilers	Solid	1.00	0.05	0.01	19420.00	1010	132
Total emissions (Mg/year)						2511	572

Animal farming

Unlike arable farming, the procedure of emisions assessment from animal breeding is described in the Guidebook. Resulting emissions correspond to RAINS emissions estimates.
For emission estimation from animal farming, species composition and age structure of animals in the Czech breeds and breeding techniques were taken into account.

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Particle emissions from German livestock buildings - influences and fluctuation factors

Ch. Nannen[1] and W. Büscher[1]

Abstract

Dust measurements are done in several animal buildings with different air ventilation and feeding systems. Even if all dust regulations are observed some aspects should be added, which apply to influences on the dust emission factors. These aspects are related to mean emission factors over one day, season or year. For that the parameters air volume flow, animal activity and particle concentration in the interior and exhaust air depending on different seasons are done in a fattening pig barn. Animal activity and air volume flow affected the developing of particle emission factors. A deviation of single half an hour data to the average value of the whole day was determined. So it is necessary to set the minimum analysing interval on 24 hours. The measuring point for the dust measurements should be the exhaust air. A comparison of the interior and exhaust dust concentrations showed that fact.

Furthermore the distribution of particles is changing by different levels of animal activity. The higher the animal activity the higher is the number of particles with big optical diameters. The management of the farmer has a big influence on the interior factors of the barn. Different practises with equal housing and feeding systems, weights of animals and ventilation systems yield different results.

Keywords: dust measurement, animal activity, particle emission, ventilation rate

Introduction

In most animal houses, heating, feeding and ventilation systems are very complex and have many influences on the generation of particles, odor and ammonia. Based on several measurements on dust we are able to describe some important conditions for the measurement setups. The influences of animal activity and ventilation rates should be exposed to get more knowledge about the considering fluctuation factors.

Methods

Diurnal variations in the parameters animal activity, particle concentration and air volume flow rate were recorded with different weather conditions in order to facilitate the analysis of the connection between animal activity and particle emissions. The investigations were made in a fattening house for 112 pigs with dry feeding (ad libitum) and door ventilation.

The continuous particle measurements were carried out in accordance with standardised measuring instructions for occupational safety and health (VDI 2080, VDI 2066). Particle concentrations were measured with an aerosol spectrometer, model 1.108 (GRIMM-Aerosoltechnik, Ainring, Germany). Isokinetic sampling in the exhaust air flow was carried out on the intake side. The measuring interval was one minute. A calibrated measuring fan with a data logger for monitoring the frequency of the fan was used to monitor the volume flow rate. The measuring interval in this case was one minute as well. The measuring fan was positioned on the pressure side of the exhaust shaft. The activity sensors used in this study are commercial passive infrared sensors modified for this particular application. The integrated relay control for the prevention of interferences has been deactivated. Instead, a voltage is tapped directly from the sensor so that it maps a signal that is analogous to the recorded movement. The signal is a differential signal, i.e. rapid temperature increases or decreases in front of the sensor lead to higher positive or negative values than slow changes in temperature. Thus, the amplitude of the impulse is proportional to the intensity of the temperature change. Slow temperature changes induce no signal. Signals are first rectified for further processing so that temperature increases and decreases are no longer differentiated. The

[1] Section Livestock Technology at the Institute for Agricultural Engineering of Bonn University, Germany

incoming signal is then routed through a holding circuit so that short impulses are artificially prolonged; the duration can be adjusted on the sensor. Thus, a data logger records the voltage every six seconds to capture short movements as well.

To get more knowledge about the difference of the particle concentration between the interior air and the exhaust air, two aerosol spectrometers were positioned inside the barn 1.2 m above the animals and in the exhaust air as described before.

Results and discussion

The first and most important fact is that it is necessary to measure emissions over one day to calculate a mean value for those emissions. The deviation of a three or four hour's measurement is factor 2 to 4 in relation to the daily mean. Figure 1 shows that fact.

The reasons for this deviation are changing animal activity and changing air volume rates. So it is possible to underestimate or overestimate the emissions by measuring only short terms.

Animal activity and particle emission levels are closely correlated. Emissions increase drastically especially during periods of activity as you can see in figure 2. The results of the measurements by Pedersen S. (1993) and Bönsch S. et al. (1996) were confirmed by the measurements made for the present study. The example of the development of animal activity and particle emission for several days as depicted in figure 3 shows that especially surveillance walks are connected with high animal activity and consequently with high particle emission. The question arises to what degree animal activity and particle emission are correlated. Figure 3 also contains the scatter plot of the correlation. As expected, the calculated coefficient of determination across all size classes (R_2 = 0.8) shows the correlation between animal activity and particle emission to be high.

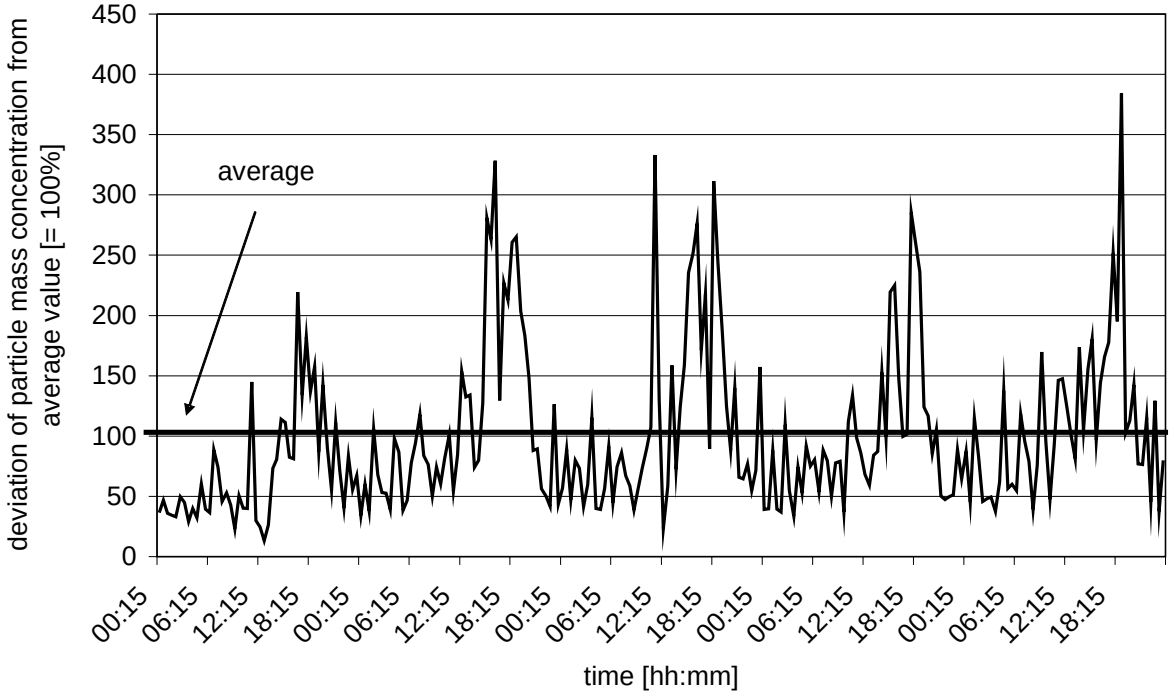

Figure 1:
Deviation of half an hour values of particle mass concentration from average value

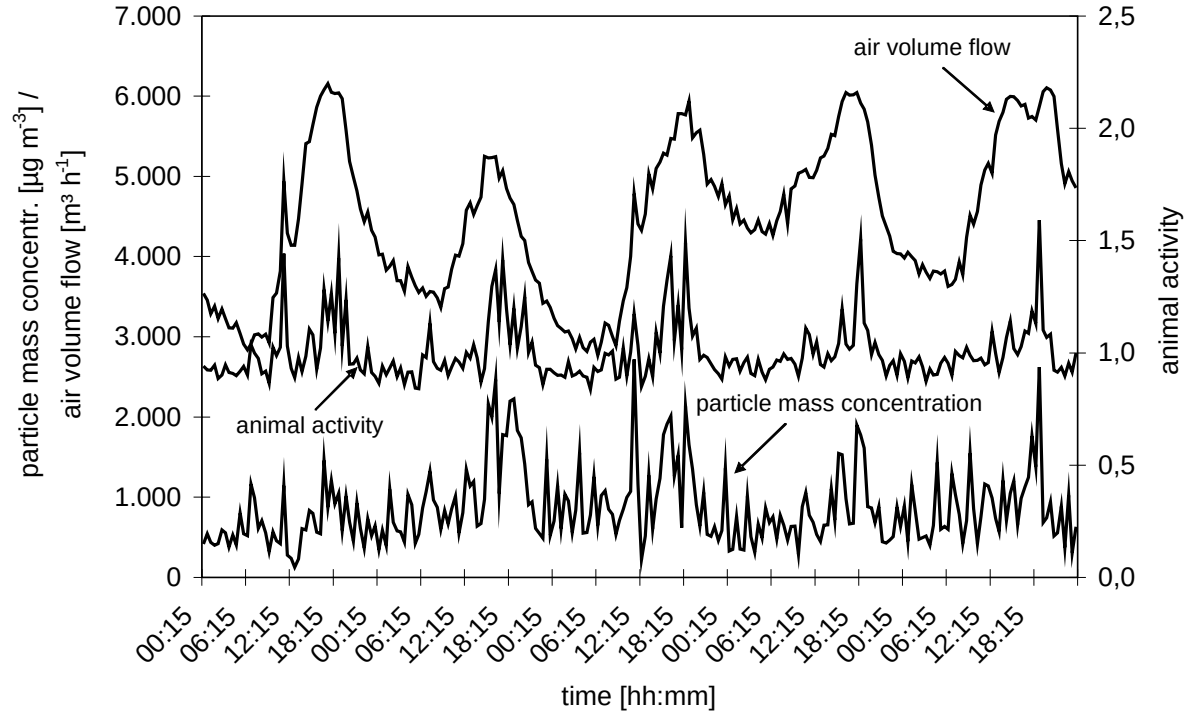

Figure 2:
Particle mass concentration, air volume flow and animal activity

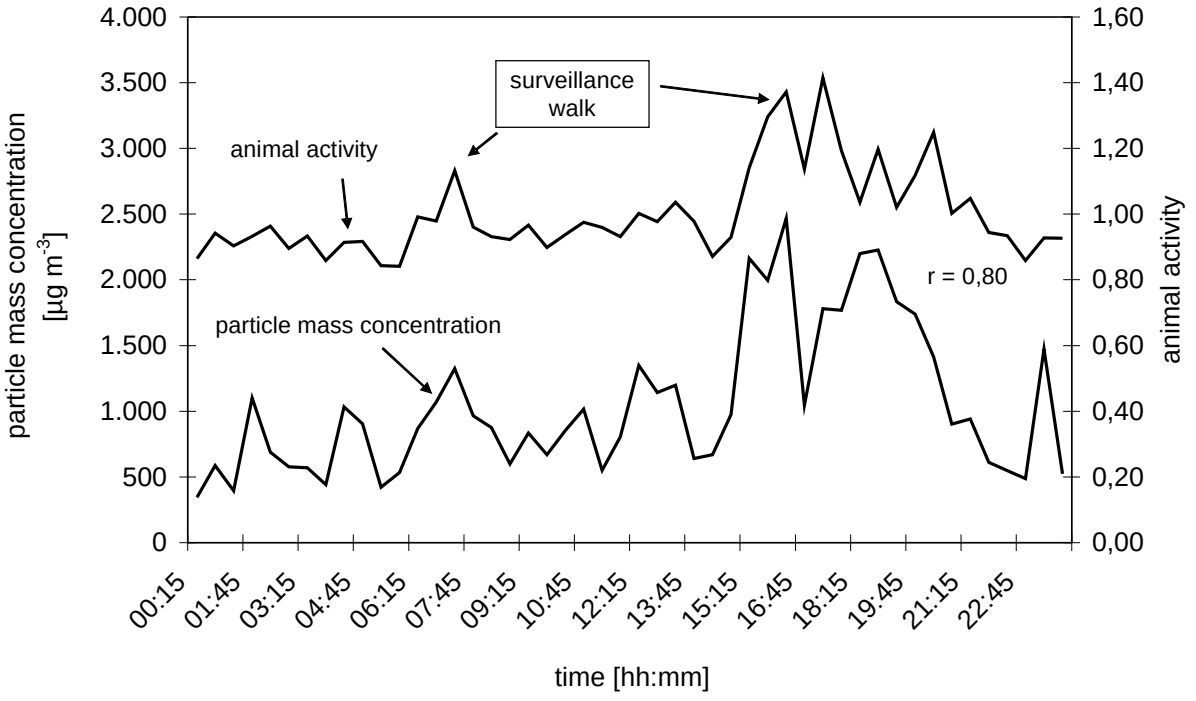

Figure 3:
Particle mass concentration and animal activity - relationships

In this analysis a new aspect was found regarding particle size distribution. Representative for all daily developments, figure 4 shows the development of the total mass concentration, the PM10 concentration and the levels of animal activity.

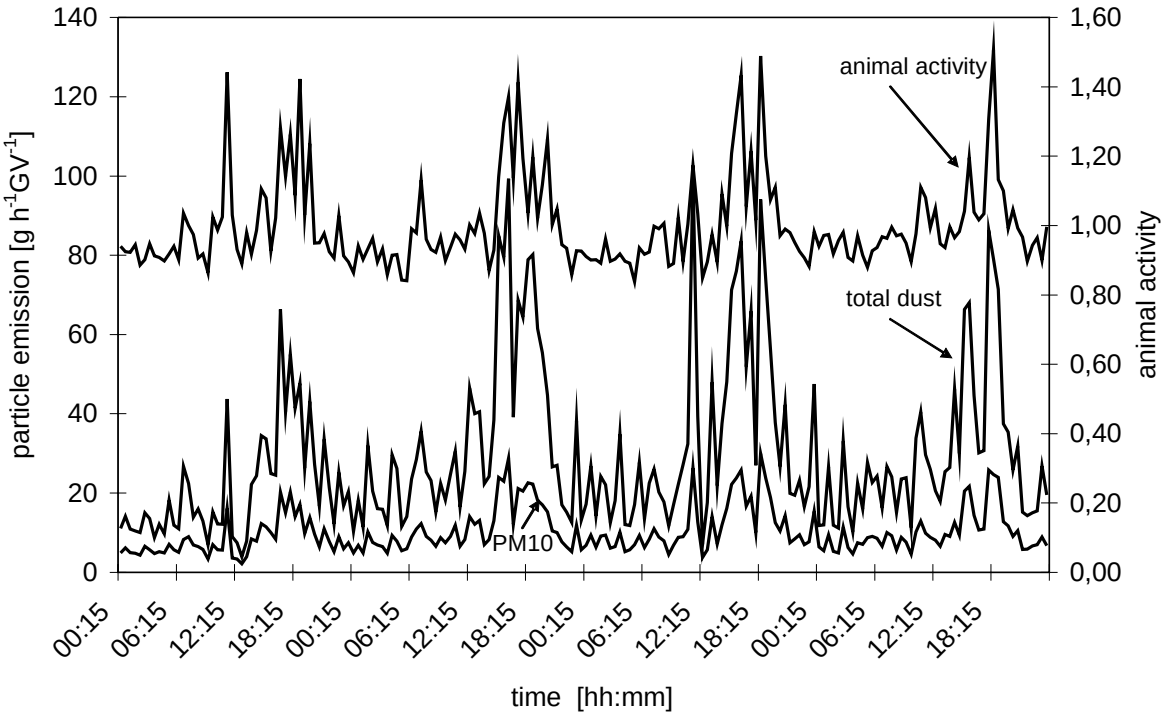

Figure 4:
Total particle mass concentration and PM10 concentration in relation to animal activity

One of the reasons for this is the increasing proportion of larger particles in the total number of particles. In this case high animal activity describes a deviation of one standard deviation from the average value of the day set 100 %. Higher levels of animal activity affect especially the particles with increasing diameter.

The higher the ventilation rate, the lower the dust concentration and the higher the correspondence between the dust concentration in the interior air and in the exhaust air due to better mixing. Because of that, it is necessary to measure the emission in the exhaust air and not in the barn. We calculate factors between the interior and exhaust air depending on different levels of the air volume flow. It was concluded that the barn is not a constant system that emits equal values of emissions over the whole day. The higher quotients for particles >5 µm can be attributed to the deposition of dust in the fan shafts. Particles are deposited on the walls of the exhaust air chimneys, resuspending at sufficiently high air velocities as larger particles formed by agglomeration with other particles. To visualise the results, the quotient of the particle concentration in the exhaust air and the particle concentration in the interior was calculated for different particle sizes. This concentration ratio describes the probability of encountering a particle of a certain size both on the inside of the livestock house and in its exhaust air. At a quotient of q = 1 for all particle size classes, all particles from the interior of the livestock house would make their way into the exhaust air. The particle concentration inside the livestock house would be the same as that in its exhaust air. The assumption that particle concentrations in the interior of livestock houses differ from those in their exhaust air was confirmed by reproducible measurements. The number of bigger particles in the interior air is much higher than in the exhaust air as you can see in figure 5.

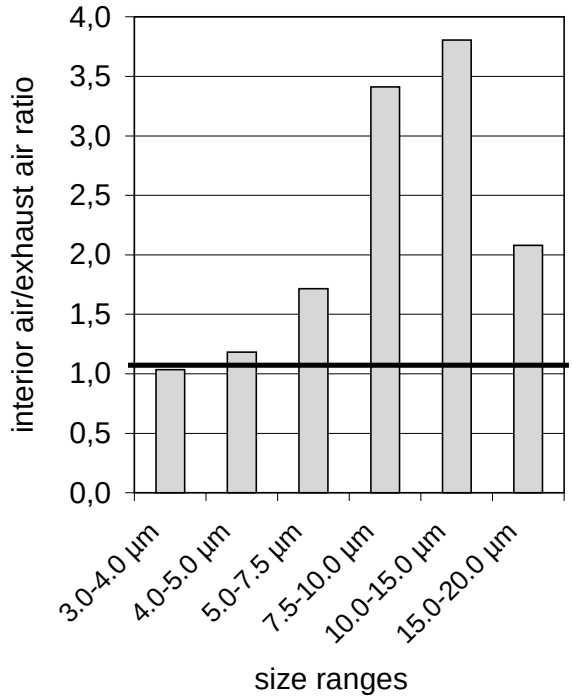

Figure 5:
Ratio of the particle number concentration in the interior air to the exhaust air

The last influencing factor is the management. Two barns, constructed in the same way with equal feeding, ventilation and heating systems were compared. Surveillance walk, adjustment of the air volume flow or number of feeding time per day affect different emission factor, expressed in emission per animal unit. Figure 6 shows the particle mass concentration and the air volume flow in two different fattening pig farms. The management of the farmer is determined by the air volume flow. The airflow is a function of the temperature set of the controller. This includes influences on the particle mass concentration and the particle emission factors.

Other important factors on emissions are the seasonal effects. Because of higher volume flow rates per animal in summer, the emissions are higher than in autumn or winter. On the other hand, the concentration of emissions in the exhaust air in the winter time is higher than in the summer time as you can see in figure 6.

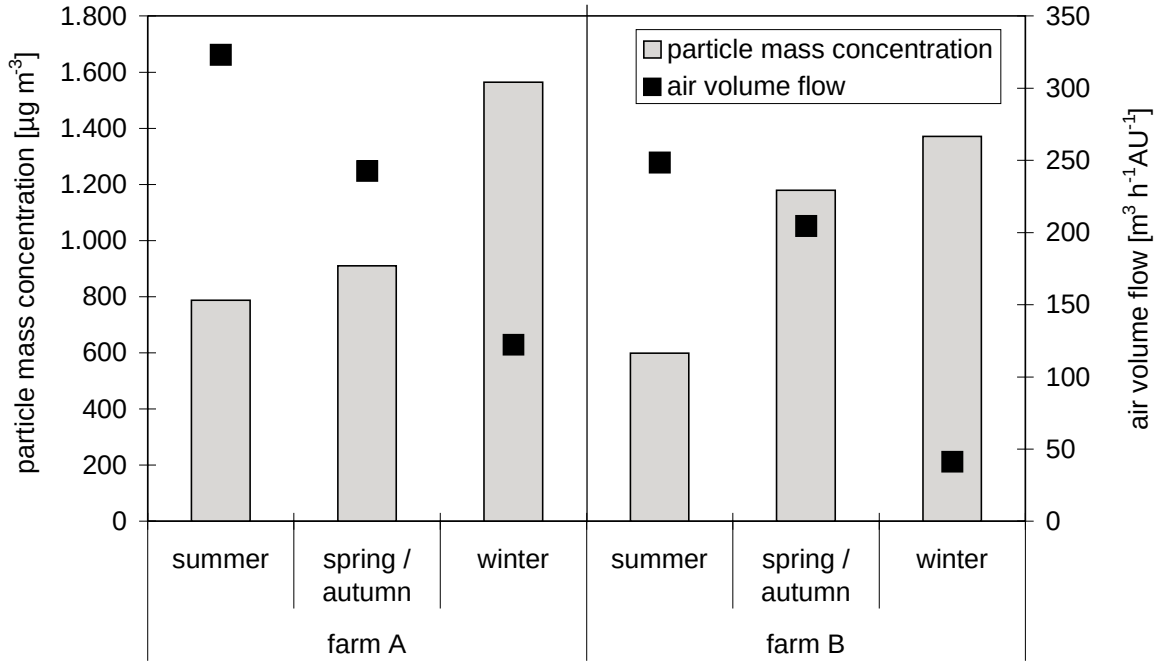

Figure 6:
Particle mass concentrations and air volume flows in different seasons and two different farms

Consequences

The results showed different considering fluctuation factors on dust generation and PM-emissions. To quantify a dust emission factor, it is necessary to measure 24 hours. In that case animal activity is the most important factor. Climatically effects and the house management of the farmer lead to differ between seasons. The position of dust sampling should be in the exhaust air, because the dust concentration in the interior air is not equal to the concentration in the exhaust air.

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Physical aspects of aerosol particle dispersion

E. Rosenthal[1], W. Büscher[1], and B. Diekmann[2]

Abstract

The dispersion of dust particles gives us the causal connection between particle emission and immission. Due to the fact that it is impossible to follow the track of a single particle through the air, a lot of effects and there parameters must be known to simulate the dispersion.

The usage of tracer-gases allows to compare (gas) dispersion models with measured data, but the dispersion of gases or odours is not comparable with the dispersion of aerosols. One reason for that is a factor 10 to the power of 10 in the weight of a one micron particle with the density of 1000kg/m³ and a SF6 molecule.

The best available technologies to determine the parameters of all participated effects are laboratory-confirmed experiments. At the Institute of Agriculture Engineering at Bonn University such experiments has been made and will be done further, supported by the DFG (German Research Foundation).

In our research we examine dust samples with agriculture background, mostly from animal farms. One major effect, which influences the dispersion of aerosols, is the sedimentation. The results of our experiments are that the sedimentation velocity and the aerodynamic behaviour of aerosol particles depends on the animal type, the feeding system and the animal house setup. Another important effect is the agglomeration, which is depending just as well on the dust source and meteorological parameters, basically from the humidity. Furthermore aerosol particles can interact with surfaces. Depending on the surface structure, the humidity and the particle attributes as well as the air velocity nearby the particle surface interface, the aerosol particles can be accumulated or displaced.

Difficulties occur in statistic dispersion simulations including all before described effects. At our institute a numerical dispersion model is developed. Every single track of all particles is computed, so that all participated physical effects which influence the behaviour of aerosol particles can be taken into account.

Keywords: aerosol transmission, dispersion models, sedimentation, agglomeration, adsorption, resuspension

Introduction

The term “aerosol transmission” describes the transport of dust particles from the emission source to the immission point. During the transmission phase the emitted substance is subject to a number of processes which change the properties and the concentration of the substance in the air. Apart from the influence of chemical decomposition, the concentration of a substance at the immission point is primarily determined by physical effects and dilution during dispersion. The dispersion of dust particles differs in important respects from the dispersion of gases. Dust particles are subject to a variety of physical processes according to their density, size and shape. The most important physical effects in this context are sedimentation, agglomeration and aerodynamics as well as adsorption and resuspension. Statements about the transmission behaviour of an aerosol require particle-specific analysis of the dust in question.

Sedimentation

It is primarily the sedimentation effect, i.e. the influence of gravity on aerosol particles, that causes them to fall to the ground. The sedimentation velocity depends not only on the weight of the particle but also on meteorological parameters (temperature, air pressure, air humidity) and, of course, on the size of the particle. This is illustrated in figure 1, which shows the sedimentation velocities of different size classes in a dust sample from an aviary house with approximately 70,000 laying hens (Schmitt-Pauksztat G. 2006).

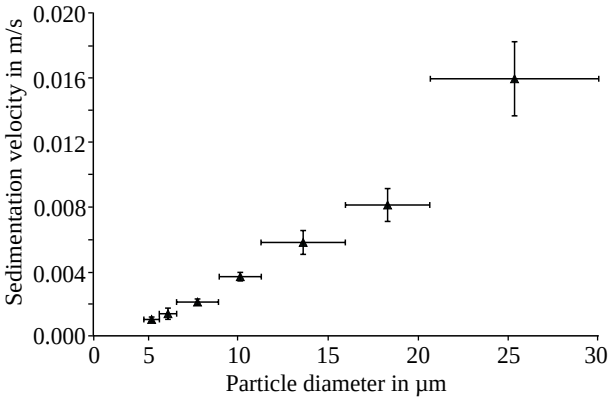

Figure. 1:
Measured sedimentation velocities of different particle size classes in a dust sample from an aviary house for laying hens (Schmitt-Pauksztat G., 2006)

[1] Institute of Agricultural Engineering, Bonn University, Germany
[2] Institute of Physics, Bonn University, Germany

Measured sedimentation velocities and particle sizes can be used to calculate the ratio of particle density to particle shape. Figure 2 shows that the ratio of the density to the shape of the particles of the dust sample used in this example differs according to their diameter. In the same sample, the ratio is three times higher for small particles than for large particles.

A possible explanation for the large differences between the individual particle sizes is not so much the variance of the particle densities as the different shapes of the particles. The density of smaller particles with a diameter of 3-5µm is in the same range as concrete, which has a density of 2000 to 3000 kg/m³ depending on the admixtures used, whereas the density of larger particles is in the range of organic material such as feed fines, epidermal scales or feather fragments. Moreover, in mineral particles the form factor is usually smaller than in organic particles so that the denominator and the numerator can lower or raise the ratio at the same time. Therefore, it is possible to gain information on the possible origin of dust particles by measuring the sedimentation velocity of a dust sample and calculating the ratio of particle density to particle shape.

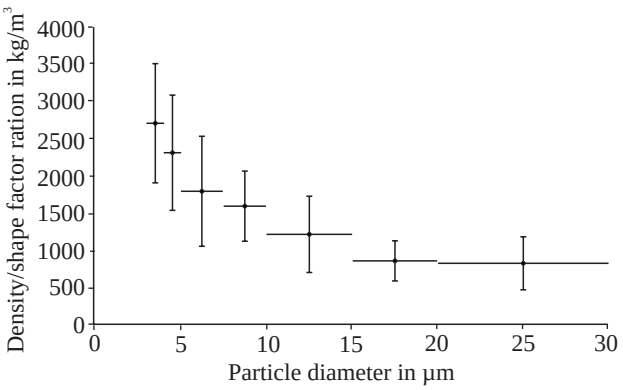

Figure 2:
Ratio of particle density to particle shape factor for different particle size classes in a dust sample from an aviary house for laying hens, calculated from experimental data

Agglomeration

Another important effect in relation to the transmission of aerosols is agglomeration. Agglomeration is the association of individual aerosol particles in so-called agglomerates as a result of attractive, binding forces. As the range of the forces between individual aerosol particles is very limited, agglomerates are formed almost exclusively as the result of two particles colliding. The collision of particles due to their thermal movement is called thermal coagulation. The binding forces between particles are mainly electromagnetic and hydrostatic forces. Differing from other particles in terms of diameter and shape, aerosols also have different aerodynamic properties and different sedimentation velocities. Experiments have shown that air humidity in particular has a great influence on the agglomeration behaviour of aerosols. Due to electrostatic forces the agglomeration rate is high at low air humidity, whereas hydrostatic forces stimulate agglomeration at high air humidity. Moreover, agglomeration depends on the material properties of the particles so that the variables describing the agglomeration process may vary for different aerosols from different kinds of sources (Rosenthal E. 2006).

Particle shape

Another important parameter in aerosol dispersion is the geometry or the shape of the particles. Microscopic studies show that dust from agricultural sources consists of highly structured particles of various shapes and densities (fig. 3).

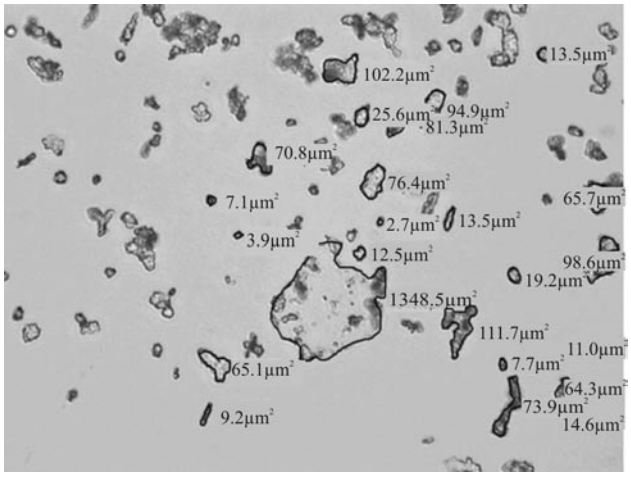

Figure 3:
Microscopic image of aerosol particles; the figures next to the particles specify their respective size (Nannen C., 2005)

The aerodynamic behaviour of a particle is influenced by its shape in the same manner as that of a car. To do justice to the various surface shapes of particles, a number of approaches to describing the aerodynamic properties of particles have been developed. The dynamic shape factor is a parameter that is used to adapt the Stokes friction term for irregularly shaped particles. Very much like the Cd factor (cars), the shape factor is very difficult to calculate and is therefore usually determined by experiments. Another way of adapting the Stokes term is by using the aerodynamic diameter, which is defined as the diameter of a spherical particle of unit density (1000 kg/m³) which has the same sedimentation velocity as the irregularly shaped particle under consideration (Hinds W. C. 1992). The Stokes diameter, which is used less frequently than the aerodynamic diameter, is the diameter of a spherical particle that has

the same density and the same sedimentation velocity as the irregularly shaped particle under consideration. With a procedure developed at the Institute for Agricultural Engineering of Bonn University the ratio of particle density to particle shape can be determined experimentally. This measuring procedure is based on a comparison of experimentally determined sedimentation velocities with the corresponding optically measured size classes. Thus, the aerodynamic behaviour of particles of any shape can be taken into account in transmission simulationss (Schmitt-Pauksztat G. 2006). Analyses of the dispersion behaviour of aerosols must also take into account the interface behaviour between aerosol particles and various kinds of surface. The process of particles settling on a surface is called adsorption. The process of particles being detached again from the surface by airflow is called resuspension. Adsorption and resuspension both depend on the surface structure (roughness, physical and chemical properties) and on the properties of the particle under consideration. For precise predictions of the dynamic dispersion of aerosols it is necessary to parameterize the adsorption and resuspension processes with suitable variables, which have to be determined experimentally.

Immission and dispersion modelling

By means of flow simulations based on the physical effects described above and on parameters which have to be determined experimentally in advance, it is possible to analyse and predict the dynamic distribution of aerosols in the exhaust plumes of livestock houses. Dispersion simulations are often based on statistical plume models, the main advantages of which are short computing times and low computing performance requirements. Moreover, such models produce reliable predictions for areas further away from livestock houses, provided that the ground is level and that the meteorological conditions are stable. Statistical models have clear drawbacks, however, if the focus is on an area within only a few kilometres from the emission source. For example, turbulences caused by the animal houses themselves, by neighbouring buildings or by natural obstacles (trees, hedgerows, etc.) cannot, or can only insufficiently, be integrated in statistical models. This is partly due to the inertia of dust particles, which makes itself felt especially in turbulent airflows. Lagrange models seem to be more suitable for aerosol dispersion simulations. They use a stationary flow field on which the trajectories of the articles are calculated; averaged turbulences can be used as parameters in the calculation. For determining the travel distance of bioaerosols and the influence of neighbouring facilities it is important to look at the immediate vicinity of the source buildings and any turbulences that might occur there. Experimental findings by the State Environmental Agency of North Rhine-Westphalia have shown that the travel distance of aerosols depends strongly on the design and the resulting flow conditions of buildings on the immission side (Heller D. 2004). In order to be able to predict the dispersal of aerosols within a few kilometres while taking the buildings in that area into account, a numerical dispersion simulation for near-surface aerosols was developed at the Institute for Agricultural Engineering in co-operation with the Physics Institute of Bonn University (Wallenfang W. et al. 2002).

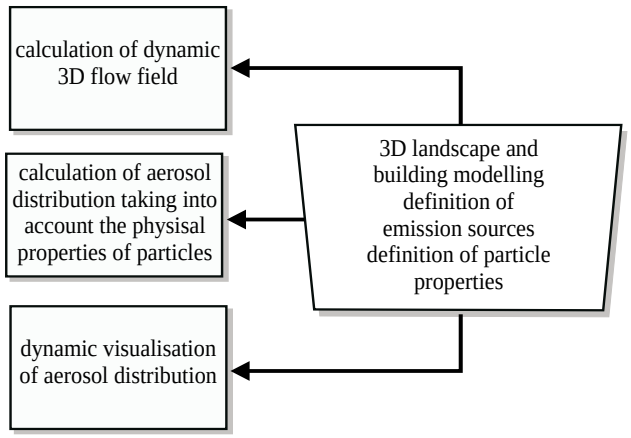

Figure 4:
Functional blocks of the NAAS 3D numerical dispersion simulation program

In a research project funded by the German Research Foundation (DFG – Deutsche Forschungsgemeinschaft) the three-dimensional aerosol dispersion simulation program (NAAS 3D – Numerische Aerosol Ausbreitungssimulation in drei Dimensionen) is being developed further. In contrast to statistical models, in NAAS 3D the trajectories of individual aerosol particles are calculated on the basis of a dynamic flow simulation. The simulation process can be divided into three functional blocks (fig. 4). In the first step, a dynamic flow vector field is created on the basis of three-dimensional information regarding buildings, the terrain and meteorological data. The relevant airflow calculations can be carried out with any meteorological or flow-dynamical simulation program with an ASCII Tecplot interface. The dispersion behaviour of aerosols in the vicinity of buildings and natural obstacles is influenced strongly by turbulences. Therefore, a program called NaSt3DGB, which was developed by the Institute for Numerical Simulation of Bonn University, is used in the simulation; it offers a numerical solution for incompressible Navier-Stokes equations (Griebel et al. 1995). In the next step, the trajectories of the aerosol particles are calculated on the basis of the dynamic vector field. This step is based on the physical parameters of the individual particles. Here the great ad-

vantage of the numerical calculation of the particle trajectories becomes evident; for instance, the agglomeration behaviour of the particles can be modelled with a high degree of precision. Moreover, the characteristic shape of each particle and the interaction between particles and surfaces are taken into account by using the aerodynamic diameter and experimentally determined variables, respectively. In the third and last functional block of the dispersion simulation the particle trajectories are visualised and the immission data are represented graphically. The representation of the aerosol distribution is generated by the computer in real time. This means that the computer continuously computes every individual image from the available geometrical data and from the behaviour and movement of the 'observer' (fig. 5). Thus, the user is provided with a three-dimensional insight into the spatial distribution of an aerosol from one or more sources in the vicinity of a building.

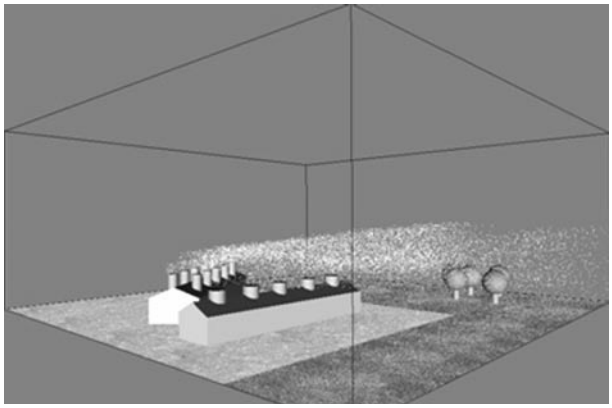

Figure 5:
Visual representation of a time step in a NAAS 3D numerical dispersion simulation

The three model variants presented in this paper can be classified as follows:

- Statistical programs
- Lagrange model
- Dynamic dispersion models

Statistical programs show their advantages in the prediction of odours if the immission point is a few kilometres removed from the emission source in less structured terrain. The Lagrange model strikes a compromise between the required calculation time and the precision of the prediction. It is only of limited use in the closer vicinity of emission sources. If the focus is on the dispersal of aerosols in complex terrain a few hundred metres around the source, then numerical calculation methods based on dynamic flow fields are the best choice.

Suitable tracer systems for the validation of aerosol dispersal simulations are currently being researched at the Institute for Agricultural Engineering. Tracer gases, such as SF6 which is used to validate odour dispersal models, are unsuitable because they do not replicate the particle-specific properties of aerosols. The difficulty in developing a suitable aerosol tracer system lies in linking measured dusts to particular emission sources. The two approaches taken to the problem of identifying dusts can be divided into online and offline methods.

The use of modified two-stage optical particle counter suggests itself fort he online approach. In the first stage, the size of a particle is measured by means of scattered light. On the basis of additional information on the number of particles it is possible to calculate the development of the concentrations of different particle size class. In the second stage, the particle counter checks if the particle can be attributed to the tracer source.

In the second approach, offline evaluation, particle samplers are used to collect the dust load together with the tracer dust at different times and at a specified distance from the source. In a laboratory the particles are examined with a light microscope in order to determine their size and distribution. One suitable kind of tracer dust can be pollen from plants which do not occur in the natural surroundings of the area under scrutiny at the time of the measurements. Fluorescent particles, which are easy to identify under the microscope in suitable light conditions, are another option, and they are also suitable for use in online methods.

Yet another option is the detection of released radioactive nuclide (Müller H.-J. 2001). Being much more sensitive than methods based on tracer particles, this method requires only small amounts of radioactive material to be released. Suitable radionuclides are pure gamma emitters with short half-life periods in the range of a few hours. Due to its environmental impact, however, this method is suitable only for sporadic research and validation measurements and not for use on a routine basis.

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Human health risks from diesel engine particles

J. Bünger[1]

Summary

Evidence from toxicological and epidemiological studies indicates that human health hazards are associated with exposure to diesel engine particles (DEP), also called diesel particulate matter (DPM). The hazards include acute exposure-related symptoms, chronic exposure related non-cancer respiratory effects, and lung cancer (EPA 2002). As new and cleaner diesel engine and fuel technology, together with efficient exhaust aftertreatment, replace a substantial number of older engines, the general applicability of the health hazard conclusions will need to be reevaluated. With new engine and fuel technology expected to produce significantly cleaner engine exhaust, significant reductions in public health hazards are expected for those engine uses affected by the regulations. However, every newly developed technology has to be examined according to its influence on human health risks.

Keywords: diesel engine emissions, particles, polycyclic aromatic hydrocarbons, health risks, lung cancer, cardiovascular diseases, asthma

Characterization of DEP

The combustion of diesel fuel forms a complex mixture of hundreds of organic and inorganic compounds in the gas and particle phase of diesel engine emissions (DEE). Toxicologically relevant compounds in the gas phase of DEE – but not discussed in this review - are carbon dioxide, nitrogen oxides, carbon monoxide, sulfates, aldehydes (formaldehyde, acetaldehyde, acrolein), benzene, and 1,3-butadiene. Toxicologically most relevant compounds adsorbed onto surfaces of diesel particulate matter (DPM) are PAHs as well as their derivatives nitrated PAHs and oxidized PAHs. To a lesser extent (PAHs) and (nPAHs) are also found in the gaseous phase of DEE (Scheepers P. T. J. and Bos R. P. 1992a and 1992b, Health Effects Institute 1995).

The International Agency for Research on Cancer (IARC) classified DEE probably carcinogenic to humans (Group 2A) in 1989. This classification was confirmed by many other institutions.

Constituents

DEP contain a large variety of organic compounds adsorbed onto a center core of elemental carbon, as well as small amounts of sulfate, nitrate, metals, and other trace elements. DPM consists of fine particles (fine particles have a diameter <2.5 µm), including a subgroup with a large number of ultrafine particles (ultrafine particles have a diameter <0.1 µm). Collectively, these particles have a large surface area which makes them an excellent medium for adsorbing organic compounds. Also, their small size makes them highly respirable and able to reach the deep lung. A number of potentially toxicologically relevant organic compounds are on the particles. The organics, in general, range from about 20 % to 40 % of the particle weight, though higher and lower percentages are also reported. Many of the organic compounds present on the particle and in the gases are known for mutagenic and carcinogenic properties. Especially, PAHs, nitro-PAHs, and oxidized PAH derivatives are present on diesel particles, with the PAHs and their derivatives comprising about 1 % or less of the DPM mass (Scheepers P. T. J. and Bos R. P. 1992b, Health Effects Institute 1999).

[1] Forschungsinstitut für Arbeitsmedizin der Deutschen Gesetzlichen Unfallversicherung (BGFA), Institut der Ruhr-Universität Bochum, Bochum, Germany

Exposure

Exposure to diesel engine emissions is ubiquitous. Highest environmental exposures occur in urban areas due to heavy traffic. All occupations where diesel engine powered vehicles or diesel engine powered machinery is in use can be potentially exposed to diesel engine emissions. Occupations with strong exposures are related to agriculture, transportation, construction, mining, and engine maintenance. Actual information on prevalence of occupationally DEE-exposed people is limited. For the total workforce in the European Union the number of exposed employees was estimated three millions based on information in the CAREX database from 1990 to 1993 (Kauppinen T., et al. 2000). In Italy, 500.000 subjects were estimated to work with exposure to DEE (Mirabelli D. and Kauppinen T. 2005). According to the Federal Agency for Work and Labour (Bundesagentur für Arbeit 2005) nearly two million employees are working under a probable exposure to DEE in Germany. The following numbers of employees in Germany are supposed to be substantially exposed to DEE:

On-road professional drivers	750,000
Off-road vehicle/machinery operators	50,000
Vehicle and machinery maintenance workers	645,000
Transportation workers	450,000
Miners	35,000

A detailed list of occupations with an assumption for probability and intensity of exposures to DEE is given in the appendix of a Swedish study (Bofetta P. et al. 2001).

DPM mass (expressed as µg DPM/m^3) has historically been used as a surrogate measure of exposure for whole DEE. Although uncertainty exists as to whether DPM is the most appropriate parameter to correlate with human health effects, it is considered a reasonable choice until more definitive information about the mechanisms of toxicity or modes of action of DEE becomes available. In the ambient environment, human exposure to DEE comes from both on-road and non-road engine exhaust. A large percentage of the population also is exposed to ambient PM2.5, of which DPM is typically a significant constituent. Exposure estimates for the early to mid-1990s suggest that annual average DEP exposure in the U.S.A. from on-road engines alone was in the range of about 0.5 to 0.8 µg DPM/m^3 of inhaled air in many rural and urban areas, respectively. For localized urban areas where people spend a large portion of their time outdoors, the exposures are higher and, for example, may range up to 4.0 µg DPM/m^3 of inhaled air (EPA 2002). In Europe, existing occupational exposure levels (OELs) for DEP are based on the measurement of elemental carbon (EC). An inter-comparison showed that existing analytical procedures for the determination of diesel particulate matter at workplaces fulfil the requirements of European standard EN 482 (Hebisch R. et al. 2003).

Some 3,500 measurements of DEE at enclosed workplaces were performed in 1.070 German work sites in 1990 to 2000 (Mattenklott M. et al. 2002), comprising results for elemental carbon (EC) and total carbon (TC). Mean exposures at workplaces under roof varied between 0.018 and 0.027 mg/m^3 for EC (0.026 and 0.047 mg/m^3 for TC). The corresponding 90 %-values were 0.055 up to 0.100 mg/m^3 for EC (0.088 and 0.150 mg/m^3 for TC). Means of exposures at underground workplaces varied between 0.095 and 0.150 mg/m^3 for EC (0.165 and 0.210 mg/m^3 for TC). The corresponding 90 %-values were 0.226 up to 0.406 mg/m^3 for EC (0.373 and 0.498 mg/m^3 for TC).

Due to the high relevance of DEE in many occupations and environmentally exposed subjects very effort is put to the development of specific and sensitive biomarkers of exposure to DEE and their health effects, especially according to PAHs (Angerer J. et al. 1997, Scheepers P. T. J. et al. 2002, Seidel A. et al. 2002, Rossbach B. et al. 2007, Wilhelm M. et al. 2007).

Health effects of DEP

Diesel exhaust particles (DEP) contribute considerably to the ambient air pollution from particulate matter, which causes acute and chronic effects in mucosal membranes, in the respiratory tract including lung cancer, and in the cardiovascular system. Due to their median aerodynamic diameter (0.01 - 0.3 µm) these particles are readily inhaled and about 10 % are deposited in the alveolar region of the lung (Scheepers P. T. J. and Bos R. P. 1992b, Health Effects Institute 1995). Number and size of the particles as well as the type and amount of combustion compounds vary in a wide range depending on the engines, fuels, and driving parameters. Although the epidemiological evidence is strong, there are until now no established biological mechanisms to fully explain the toxicity of DPM to humans (Salvi S. et al. 1999).

Non cancer effects

Acute and subacute effects

First recognized during smog episodes in London during the 1950´s, subsequent epidemiologic studies have noted that short-term increases in ambient levels of particulate matter (PM) are associated with hospital admissions and deaths from cardiovascular and respiratory disorders. These acute effects were observed in patients with chronic obstructive pulmonary disease, chronic bronchitis, asthma and cardiovascular disease (Dockery D. W. and Pope C. A. III 1994, Samet J. M. et al. 1995, Katsouyanni K. et

al. 1997). However, the biologic mechanisms that underlie this association and the role that the composition and size of PM may have in causing adverse health effects are not well understood (Salvi S. et al. 1999, Holgate S. T. et al. 2003).

Recent experimental studies in healthy human volunteers showed increases of many parameters of systemic and pulmonary inflammation but no measurable effects on lung function after exposure to DEP (Salvi S. et al. 1999, Nordenhall C. et al. 2000, Pourazar J. et al. 2005). Furthermore, there is broad evidence that DEP inhalation causes vascular dysfunction and impaired endogenous fibrinolysis (Mills N. L. et al. 2005).

Information is limited for characterizing the potential health effects associated with acute or short-term exposure. However, on the basis of available human and animal evidence, it is concluded that acute or short-term (e.g., episodic) exposure to DEE can cause acute irritation of the eyes and upper airways, neurophysiological symptoms (e.g., lightheadedness, nausea), and respiratory symptoms (cough, phlegm). The lack of adequate exposure-response information in the acute health effect studies precludes the development of recommendations about levels of exposure that would be presumed safe for these effects (EPA 2002).

Chronic effects

In long-term studies a significant association of mortality has been reported with air pollution from PM less than 10 µm in aerodynamic diameter (PM10) (Schwartz J. 1993, Dockery D. W. et al. 1993, Pope C. A. III et al. 1995, Abbey D. E. et al. 1999). The Working Group on Public Health and Fossil-Fuel Combustion (1997) estimated 8 million additional deaths globally until 2020 due to PM exposure if the air pollution increases to the same degree as hitherto. Recently, special attention has been focussed on the respiratory effects of fine (PM2.5) and ultrafine particles (PM0.1) (Peters A. et al. 1997, Schwartz J. and Neas L. M. 2000).

Information from human studies is solely inadequate for a definitive evaluation of non-cancer health effects from chronic exposure to DEP. However, on the basis of extensive animal evidence, DEP is judged to pose a chronic respiratory hazard to humans. Chronic-exposure, animal inhalation studies show a spectrum of dose-dependent inflammation and histopathological changes in the lung in several animal species including rats, mice, hamsters, and monkeys (EPA 2002).

Sensitization

There also is evidence for an immunologic effect–the exacerbation of allergic responses to known allergens and asthma-like symptoms (EPA 2002). Several large epidemiological studies have demonstrated a strong association between exposure to motor vehicle traffic emissions and allergic symptoms and reduced lung function (Brunekeef B. et al. 1997, Brauer M. et al. 2002, Janssen N. A. et al. 2003, Mc Connell R. et al. 2006), although the singular results differ in detail. Laboratory studies in humans and animals have shown that particulate toxic pollutants-particularly diesel exhaust particulates-can enhance allergic inflammation and can induce allergic immune responses (Li N. et al. 2003, Siegel P. D. et al. 2004). Most of these immune responses are mediated by the carbon core of diesel exhaust particulates. However, also PAHs from DEP enhance the production of immunoglobulin E (Mastrangelo G. et al. 2003).

While the evidence for the exacerbation of immunologic effects in already sensitized subjects is good, evidence for the development of allergic sensitization from diesel exhaust particulates is less abundant. Comparisons of the prevalence of hay fever, as well as positive skin-prick tests, between citizens of former West and East Germany and between Hong Kong and China civilians, have demonstrated marked differences. Crucial variations in the level of particulate air pollution from motor vehicles in these countries may account for the observed increased prevalence of atopy. In a review from 2002 Polosa and coworkers stated that allergic susceptibility associated with diesel exhaust particle exposure is clear as mud (Polosa R. et al. 2002). Two years later Heinrich and Wichmann wrote that the evidence for an increased risk for asthma and hay fever is still weak but seems to be strengthened a little (Heinrich J. and Wichmann H. E. 2004).

Cancer and mutagenic effects

Cancer

An association of increased risks for lung cancer associated with exposure to DEE has been observed in the vast majority of over 30 epidemiologic cohort and case control studies published in the literature. Distinct populations of occupational groups were studied, including railroad workers, truck drivers, heavy-equipment operators, farm tractor operators, and professional diesel vehicle drivers. Three meta-analyses confirmed the relationship of diesel exhaust exposure and lung cancer (Cohen A. J. and Higgins M. W. P. 1995, Bhatia R. et al. 1998, Lipsett M. and Campleman S. 1999).

Risk assessment data based on actual exposures to DEE was not available. Elevated risks were estimated in epidemiological studies on the basis of historical exposure scenarios. The Cohen and Higgins report (1995) is a qualitative review of 35 epidemiologic studies (16 cohort and 19

case-control) of occupational exposure to DEE published between 1957 and 1993. The evidence suggests that occupational exposure to DEE from diverse sources increases the rate of lung cancer by 20 % to 40 % in exposed workers in general, and to a greater extent among workers with prolonged exposure. Bhatia R. et al. (1998) found a small but consistent increase in the risk for lung cancer among workers with exposure to DEE, in a quantitative meta-analysis of 23 studies. The pooled RR weighted by study precision was 1.33 (95 % CI = 1.24-1.44). Lipsett and Campleman (1999) performed a quantitative meta-analysis. Pooled RRs for all studies and for study subsets were estimated using a random effect model. A pooled smoking-adjusted RR was 1.47 (95 % CI = 1.29-1.67). Although the results of these analyses were called into question by several authors (Stöber W. and Abel U. R. 1996, Muscat J. E. 1996, Crump K. S. 1999, Hesterberg T. W. et al. 2006), another recent study added new evidence for an increased lung cancer risk from exposure to DEP (Parent M. E. et al. 2007).

Risk assessment data for specific occupations based on actual exposures to DEE was not available. A pooled study based on exposures in East and Western Germany some decades ago showed the following odds ratios (Brüske-Hohlfeld I. et al. 1999):

Heavy equipment operators
OR = 2.31 (95 % - CI 1.44 - 3.70)
Tractor drivers (exposure >30 years)
OR = 6.81 (95 % - CI 1.17 - 39.51)
Professional drivers (West Germany)
OR = 1.44 (95 % - CI 1.18 - 1.76)
Other traffic related jobs
OR = 1.53 (95 % - CI 1.04 - 2.24)

Most assessments conclude that DEP is “likely to be carcinogenic to humans by inhalation” and that this hazard applies to occupational and environmental exposures (IARC 1989, EPA 2002). This conclusion is based on the evidence from human, animal, and other supporting studies. There is considerable evidence demonstrating an association between DEP exposure and increased lung cancer risk among workers in varied occupations where diesel engines historically have been used. The human evidence from occupational studies is considered strongly supportive of a finding that DEP exposure is causally associated with lung cancer, though the evidence is less than that needed to definitively conclude that DEP is carcinogenic to humans. There is some uncertainty about the degree to which confounders are having an influence on the observed cancer risk in the occupational studies, and there is uncertainty evolving from the lack of actual DEP exposure data for the workers (EPA 2002).

In addition to the human evidence, there is supporting evidence of DEP’s carcinogenicity and associated DEP organic compound extracts in rats. The experiments showed a consistent dose-dependant incidence of lung tumors after a chronic exposure to high concentrations of DEE (Heinrich U. et al. 1986, Ishinishi N. et al. 1986, Iwai K. et al. 1986, Mauderly J. L. et al. 1987). But these results were criticized since concentrations led to an “lung-overload” with particles in the animals (EPA 2002).

Mutagenicity

Other supporting evidence includes the demonstrated mutagenic effects of DEP and its organic constituents. A high mutagenic potency of extracts of DEP was described by Huisingh J. et al. (1978) using the Salmonella typhimurium/mammalian microsome assay (Ames-Test) and has been confirmed in multiple further studies (Clark C. R. and Vigil C. L. 1980, Claxton L. D. and Barnes H. M. 1981, Belisario M. A. et al. 1984). The direct-acting mutagenicity of DEP is ascribed to substituted PAHs, e. g. nitro-PAHs (Wang Y. Y. et al. 1978, Pedersen T. C. and Siak J. S. 1981, Ohe T. 1984), while not substituted PAH require metabolic activation by S9 (IARC 1983). In recent years renewable sources, especially vegetable oils, were used to create biogenic diesel fuels. According to results using the Ames-Test, DEP extracts of plant oil derived methyl esters e.g. (SME, RME) are less mutagenic (Bagley S. T. et al. 1998, Bünger J. et al. 1998, Bünger J. et al. 2000a, Bünger J. et al. 2000b, Bünger J. et al. 2006) compared to common diesel fuel, while the combustion of crude rape seed oil resulted in an extreme increase of mutagenicity in an actual study (Bünger J. et al. 2007).

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An assessment of time changes of the health risk of PM10 based on GRIMM analyzer data and respiratory deposition model

J. Keder[1]

Abstract

PM10 particles are considered as one of the most problematical pollutants affecting the human health, especially in the urban environment, where they are emitted primarily from the mobile sources. Particles entering human respiratory system are subjected to number of deposition mechanisms and the fraction deposited in different parts of respiratory tract depends on their aerodynamical diameter. Thus, identical PM10 concentrations measured by monitoring stations may cause different health effects depending on size distribution of particles whose concentration was detected.

Synchronous measurements of suspended particles fractions PM1, PM2.5 and PM10 were provided using GRIMM Model 180 analyzer in Prague (Czech Republic capital) and in Ostrava industrial region. These enable to assess the proportion of contribution of different fraction to the total PM10 concentration. Collocated PM10 concentration data gathered by the radiometric method was available as well.

The model developed by the International Commission on Radiological Protection (ICRP) to predict the particle deposition in human respiratory tract was applied at the measured data and possible particulate dose assessment has been found for different locations and time periods.

Keywords: PM10, size distribution, health effects, GRIMM analyzer

Introduction

Particles are considered as one of the most problematical pollutants affecting the human health. They comprise typical air pollution burden especially in the urban environment, where they are emitted primarily from the mobile sources and in the industrial regions as well. Health risk depends by particulate matter concentration and particles size, morphology and chemical composition (Pope III. C. A. and Dockery D. W. 2006). The air quality standards regulate PM10 concentrations first of all. In recent years, however, the PM2.5 and PM1 particles, depositing in the lower respiratory tract and being assumed as the main cause of increased morbidity and mortality among population, were studied with the increased intensity.

Methodology and data used

Measured data from GRIMM Model 180 analyzer (GRIMM Aerosol Technik, 2003), gathered at two places in the Czech Republic during the period September 2005 – October 2006, were analyzed. The GRIMM analyzers were installed at automatic monitoring stations located in Prague (Czech Republic capital) and in highly industrialized town Ostrava. PM10 concentrations measurements by means of Thermo ESM Andersen FH 62 I-R analyzers are provided at both stations at regular basis. Parallel meteorological data such as air temperature and humidity, wind and global solar radiation are available as well at these sites. The analysis has been provided for the hourly means of all measured quantities.

Particles suspended in the air enter human body by breathing. These particles include natural materials such as bacteria, viruses, pollens, sea salt, road dust as well as anthropogenic emissions. The hazard caused by these particles depends on their chemical composition as well as where they deposit within human respiratory system. Hence, the understanding of aerosol deposition in human respiratory system is critical to human health so that the deposition of “bad” aerosol can be reduced (http://aerosol.ees.ufl.edu).

The respiratory system is works essentially as a filter. The viscous surface of the airway walls almost guarantees the deposition without re-entrainment when a particle is in contact with it. The most important mechanisms are impaction, settling, diffusion and interception. A particle entering respiratory system is subject to all the deposition

[1] Czech Hydrometeorological Institute, Praha, Czech Republic

mechanisms mentioned previously. The actual deposition efficiency of a given particle size has been determined experimentally. Several models have been developed to predict the deposition based on experimental data. A widely used one was developed by the International Commission on Radiological Protection. For the purpose of this model, respiratory system is divided into 3 parts: head airways (HA), tracheobronchial region (TB) and alveolar region (fig. 1).

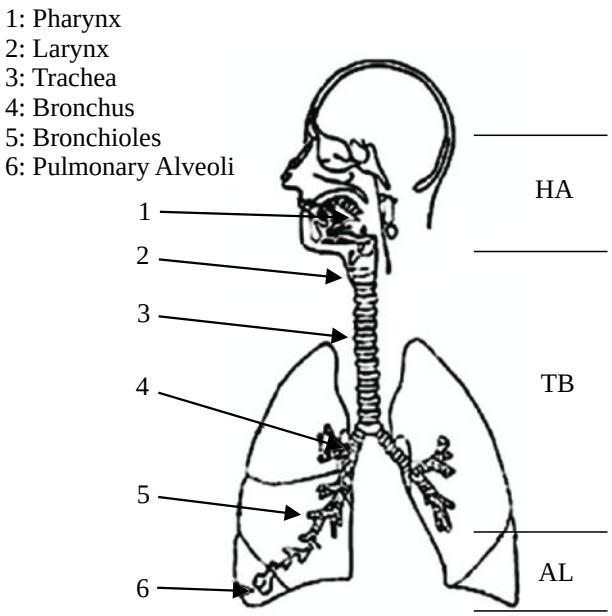

Figure 1:
Parts of human respiratory tract used in ICRP model

Equations describing the total deposition fraction (DF) in the whole respiratory system and regional deposition fractions in head airways (DFHA), tracheobronchial region (DFTB) and alveolar region (DFAL) were derived in this model (http://www.icrp.org). Regional deposition is of more interest because it is more relevant in assessing the potential hazard of inhaled particles.

In figure 2, dependences of particular deposition fraction on particle size diameter calculated according to ICRP model equations are depicted. It is obvious that particles with their diameter within the range 0.1 – 1 μm deposit with the lowest efficiency. The most dangerous, related to human health, are the fine particles with diameters ranging between 0.01 – 0.1 μm which are deposited in pulmonary alveoli without possibility to be removed from them for instance by cough.

Results and discussion

Hourly mean particle concentrations in fractions PM10 and PM2.5, measured by GRIMM analyzers, were compared with those from regular network.

Example of such comparison gives a scatter graph at figure 3, where comparison of PM10 data from Ostrava was depicted.

The good correlation of both methods is obvious, but GRIMM analyzer significantly underestimates PM10 mass concentration values. Similar results were obtained for locality in Prague and for the PM2.5 fraction. Obviously the

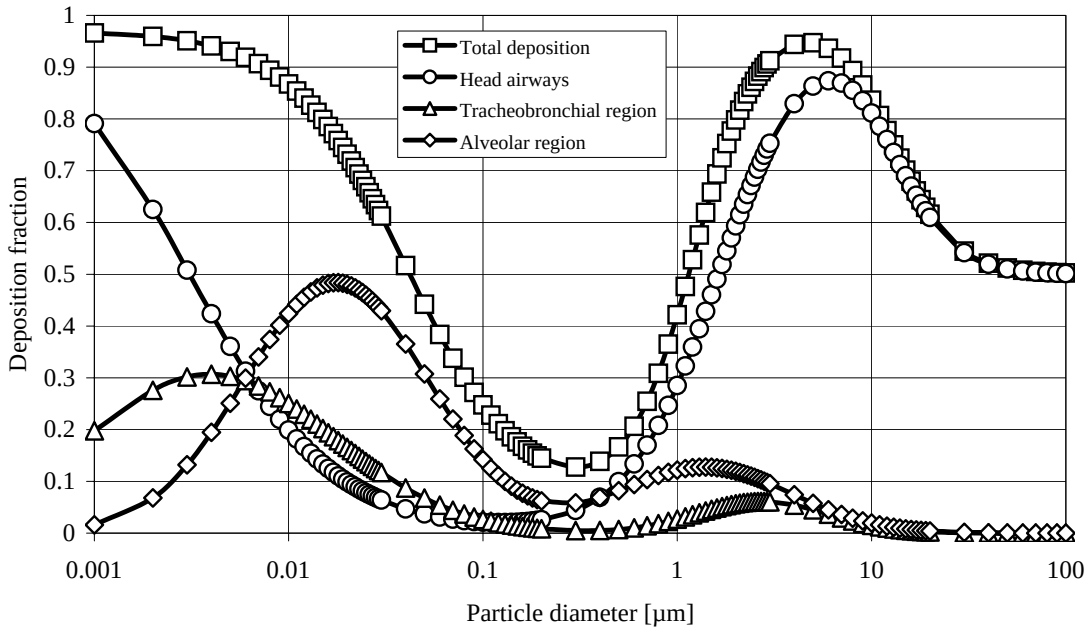

Figure 2:
Dependence of regional particle deposition fraction on particle diameter

calibration constants of GRIMM analyzer must be checked and set appropriately. As for the current data, we concluded that it would be reasonable to analyse proportions of particular fractions in PM10 instead their mass concentrations provided by GRIMM.

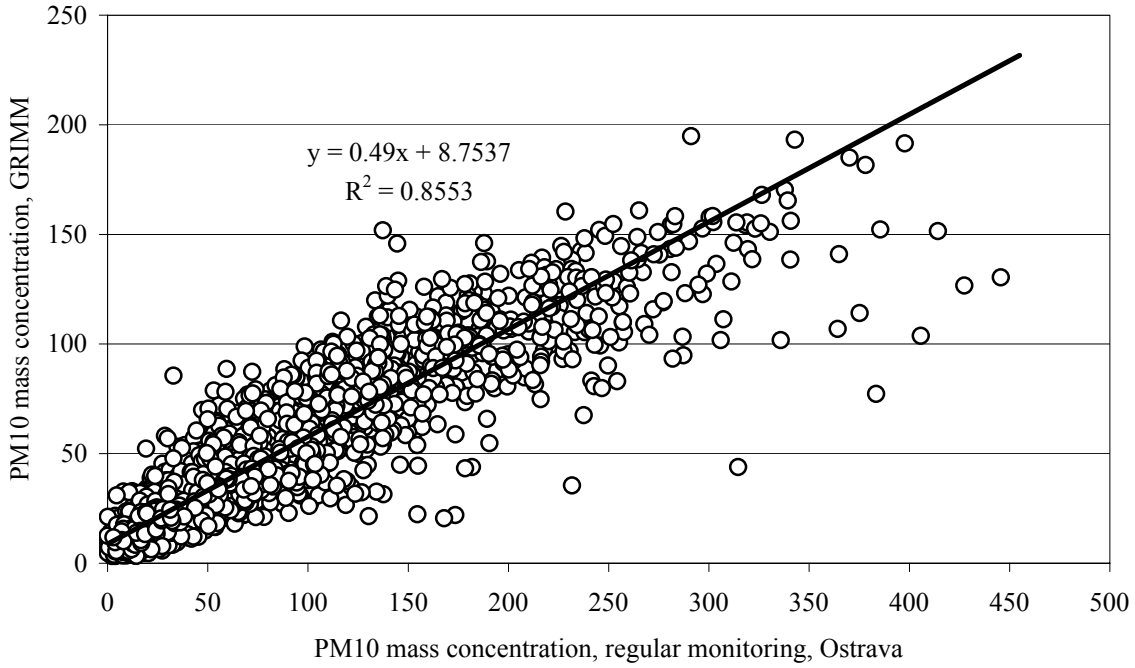

Figure 3:
Comparison of PM10 concentration measured by GRIMM and a standard network analyzer

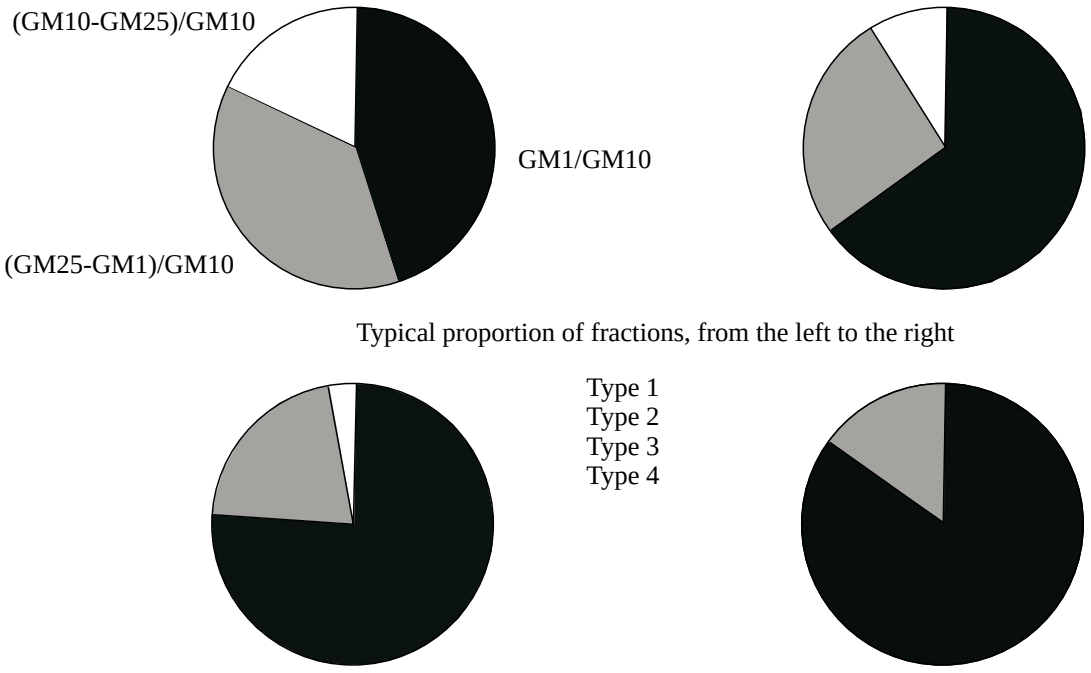

Figure 4:
Types of PM fractions proportions in PM10, Ostrava

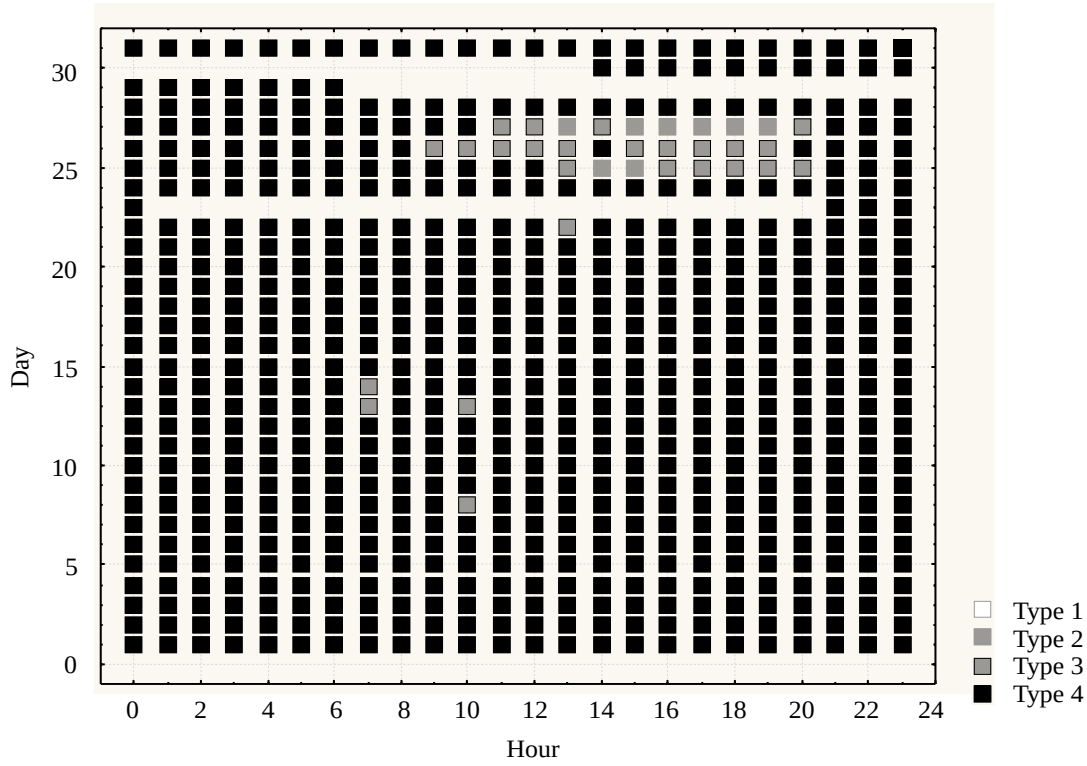

Figure 5:
Time changes of typical profiles of PM fractions share in PM10 in Ostrava, January 2006

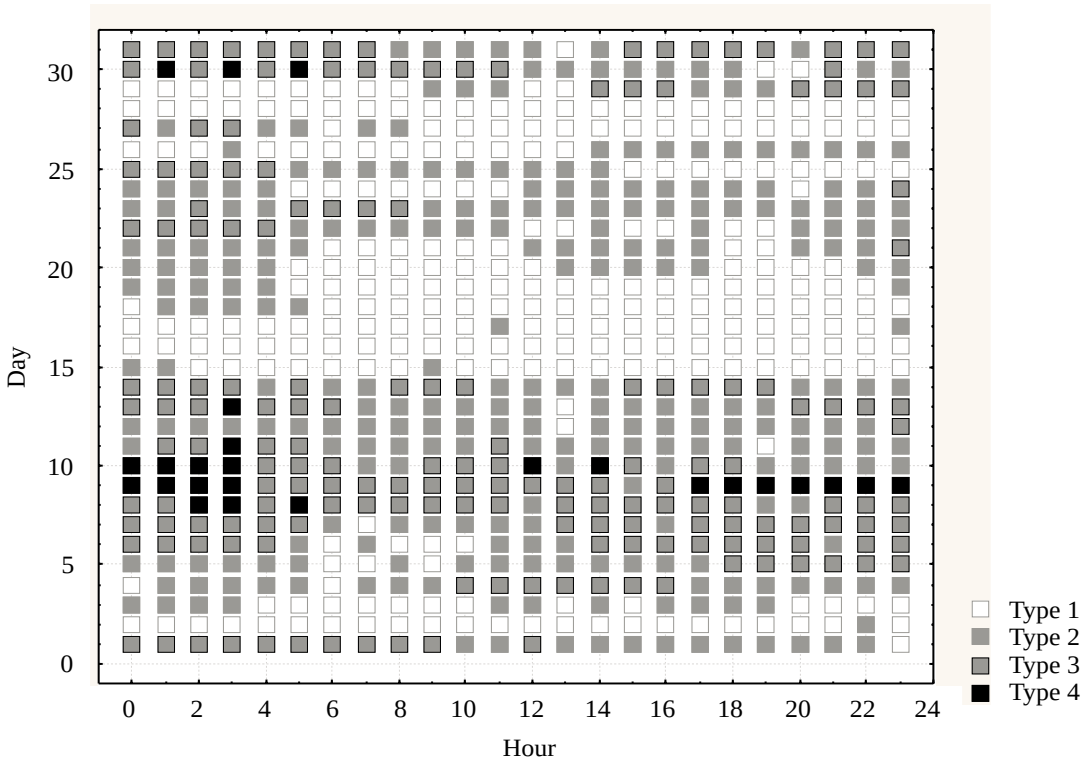

Figure 6:
Time changes of typical profiles of PM fractions share in PM10 in Ostrava, July 2006

For further analysis, proportions of particular fractions measured by GRIMM were determined, where e.g. GM1/ GM10 stays for the share of PM1 in total PM10 concentration. Changes of share of PM1 and PM2.5 in PM10 with air temperature and relative humidity were studied and it has been found that the share of fine fraction in total PM10 concentration increases when air temperature decreases. For low temperatures, practically the whole PM2.5 fraction consists of the finest PM1 particles, which are the most dangerous form the aspect of health effects. Similarly, the share of both fractions in the total PM10 increases with increasing relative humidity and the share of PM1 in PM2.5 increases as well. In range of relative humidity between 80 and 90% practically all PM2.5 consists of PM1.

For the purpose of health protection and for considerations on particle origin there is necessary to answer the question if and how the share of particular fraction in PM10 changes in time. The results of multicomponent statistical analysis showed that this share has been changing at both locations during the day and from one month to another as well. Four typical profiles of share of PM1 and PM2.5 fractions in PM10 were found by means of cluster analysis, marked from 1 to 4. Each hourly profile was then allocated into one profile class. The share of fine fraction in total PM10 increases with the increasing class number and for class 4 practically the whole PM10 consists of PM1. Thus, the rate of danger for the human health increases with the class number.

Figure 5 shows the daily changes of profile classes in Ostrava for winter (January) and summer (July) month. The most dangerous profile class dominated in Ostrava over the whole January 2006, without any indication of the daily course. The different picture showed July, when amount of less serious classes increased and daily course of them is also apparent. The described approach enables to map the changes of burden by particular PM fractions during the whole year and study their relation to meteorological parameters and emission sources types.

Clearly, not only the proportion of fractions but also their mass concentration is crucial from the viewpoint of health effects. Figure 7 gives an example of combination of parallel GRIMM and standard analyzer measurement. The PM10 mass concentration from standard analyzer has been disaggregated among particular fractions by means of coefficients derived from the share of PM1 and PM2.5 fractions measured by GRIMM. The PM1 and PM2.5 concentrations derived from PM10 ones are marked as “hypothetical”. If the GRIMM analyzer were set for measurements of broader spectrum of fractions, the parallel measurements provided by both analyzers would give the possibility to assess particulate mass concentrations in separate spectral bands and determine their time changes.

Significant changes of PM10 concentration during the time period presented at Figure 6, accompanied by simultaneous particle size spectrum changes (being demonstrated by changes of share of size fractions in PM10), caused

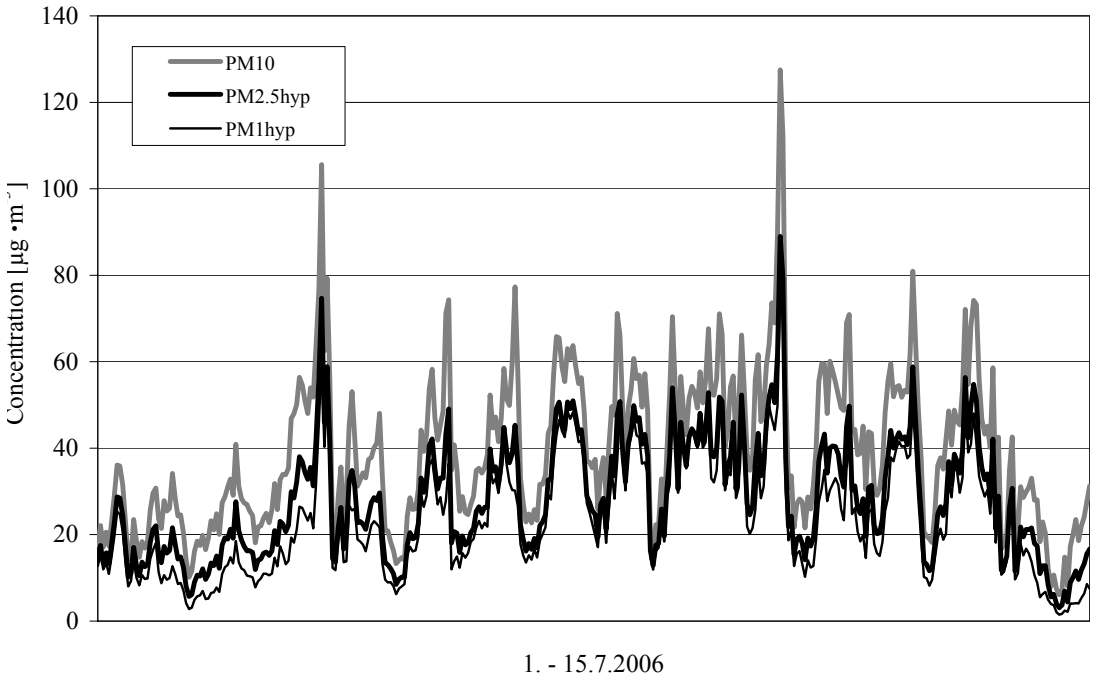

Figure 7:
Disaggregation of operationally measured PM10 mass concentrations measured by means of fraction proportions determined from GRIMM. Ostrava, July 2006

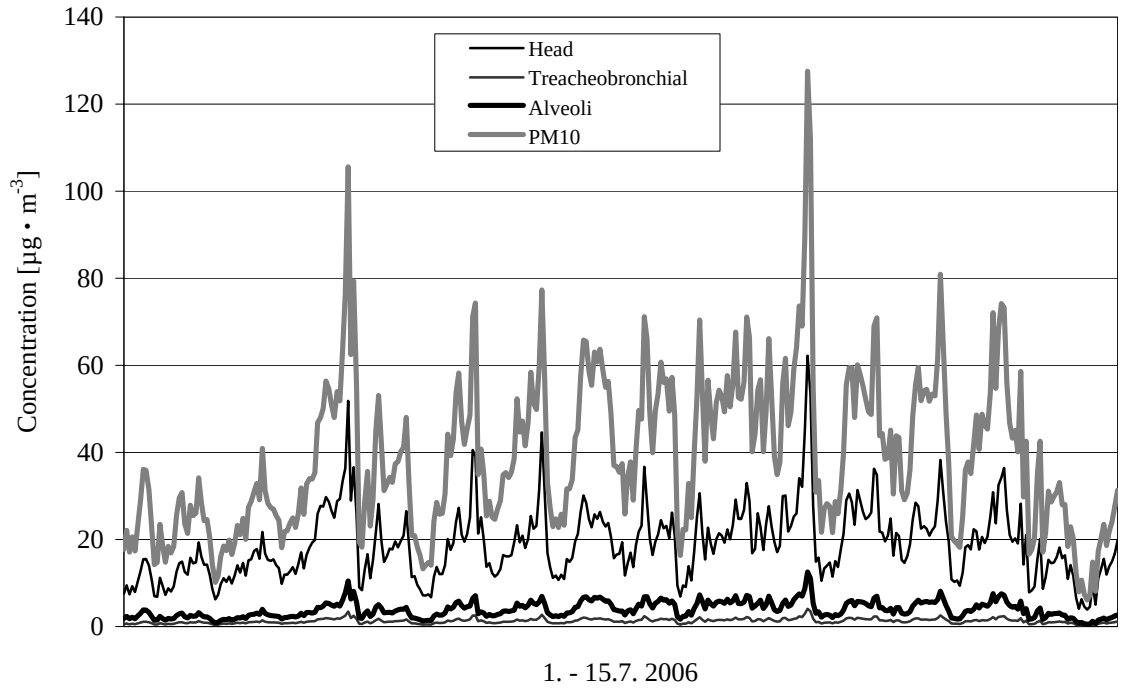

Figure 8:
Potential deposition of particle mass concentration in particular parts of human respiratory tract, derived from ICRP model

that the particle mass which might be potentially deposited in particular parts of human respiratory tract was time-dependent as well. The ICRP deposition model enables us to assess these values for each hour. Results of such assessment are shown in figure 9. Median particle diameters for each size class measured by GRIMM, i.e. 0 - 1, 1 - 2.5 and 2.5 - 10 micrometers, were taken as an input of deposition fraction calculation provided by the ICRP model. The dose potentially deposited in particular parts of respiratory tract can be treated as a measure of health hazard caused by the presence of particles in the air. This dose might differ even if the ambient PM10 concentration would be the same, depending on the actual particle size spectrum.

Conclusions

- The PM10 mass concentrations measured by GRIMM analyzer are underestimated significantly, nevertheless the data are homogenous and well correlated with the routine analyzer measurements
- It is possible to derive correction formulae for the recalculation of GRIMM data and disaggregate the PM10 data from regular monitoring into particular fractions, using the fractions proportions derived from GRIMM
- Share of particular fractions (PM1 and PM2.5) in total PM10 concentration showed significant time changes and was related to the meteorological conditions
- It was possible to define typical profiles of such proportions and to highlight the presence of structures in measured data that way
- The share of particles, depositing in the low respiratory tract (PM1 and PM2.5) at the total PM10 concentration increased with the decrease of air temperature and increase of relative humidity. This is very serious fact in relation to the human health protection
- The routine measurements expanded by GRIMM data can be used as an ICRP deposition model input and assess how the health risk caused by particles changes in time.

Acknowledgements

This work has been financially supported by the Czech Ministry of Environment from granted research project VaV SM-9-86-05.

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Aerosol-spectrometers for particle number, size and mass detection

F. Schneider[1]

Abstract

The aim of this article is to give some basic fundamentals about the functional principles of aerosol spectrometers, procedures for calibration and validation of aerosol spectrometers and some application examples of actual aerosol spectrometer used for particle number, size and mass detection in agricultural or environmental applications.

Keywords: aerosol Spectrometer, particle size distribution, PM10, PM2.5, PM1

Introduction

This paper will focus on the application of light-scattering aerosol spectrometers for particle number, size and mass detection for high particle concentrations, excluding such applications for clean spaces, where the needed specifications of the aerosol spectrometers are very different, e.g. accurate particle size determination is not requested, sample volume flow is comparably high and maximum particle concentrations are very low.

The determination of particle concentration, particle size distribution and particle mass of aerosol particles is required in various application fields, e.g. chemical, pharmaceutical, agricultural, or automotive industry. During the last decade some remarkable steps in aerosol spectrometer development took place, so today aerosol spectrometers are used to obtain all this results, mentioned above.

Aerosol spectrometers

Fundamentals and detailed descriptions can be found in literature about light scattering of single particles (Van der Hulst H. C. 1957, Bohren C. F. and Hufmann D. R. 1998), basic functional principles of aerosol spectrometers (e.g. Seinfeld J. H. and Pandis S. 1998 or in German language Haller P. 1999) and different technical specifications (e.g. Baron P. A. and Willeke K. 2001 or Hinds W. C. 1998).

Light scattering of single particles

Aerosol spectrometers are based on the interaction between an incident light beam and single aerosol particles. The fist description and calculation for the elastic scattering and absobtion by aerosol particles according to the incident wavelength are the Maxwell equations (Mie G. 1908). Light scattering can be divided in three ranges, defining an optical size parameter x by $x = \pi * Dp/\lambda$, with the aerosol particles circumference $\pi * Dp$ and the incident wavelength λ:

$x << 1$: Rayleigh-scattering: particle diameter is much smaller as the incident wavelength. The intensity of the scattered light is proportional to the sixth power of particle diameter ($I \sim D_p^6$). This is typically for e.g. gas molecules interaction with solar radiation in the atmosphere, causing blue sky effect or very small aerosol particles affecting the lower size detection limit of an aerosol spectrometer.

[1] Grimm Aerosoltechnik GmbH & Co KG, Ainring, Germany

$x \cong 1$: Mie-scattering: particle diameter is about the incident wavelength. There is a strong interaction between the aerosol particle and the incident beam, depending although on particle refractive index with no simple relation between scattered intensity and particle diameter (especially for mono chromatic light, e.g. laser). This is most relevant for atmospheric aerosol particles.

$x >> 1$: Geometric Optics: light rays hitting the particle lead to reflection, refraction and absorption, rays passing the particles edge give rise to diffraction. The scattered intensity is proportional to the particle cross-sectional area ($I \sim D_p^2$). This will cause e.g. cloud optical effects with water droplets or rainbow effect and affects an aerosol spectrometers upper size detection limit for course particles.

Beside the incident wavelength and the particle diameter the (complex) refractive index of a aerosol particle defining its scattering and absorption is very important. Depending on their chemical composition and the internal mixture of aerosol particles the refractive index influences the scattering intensity of an aerosol particle. It has to be mentioned, that scattering intensity is not symmetrically around an aerosol particle, but with strong differences in scattering angles (0° = backward direction, 90° scattering or 180° forward direction).

All together, the scattering intensity given by a single particle depends on the wavelength, intensity and polarisation angle of the incident light, the detection angle of the scattered light and the diameter and complex refractive index of the aerosol particle.

So different aerosol particles generate different scattered light impulses, which can be detected to determine particle concentration and determining the intensity of the scattered light impulse with a pulse height analyser it is also possible to determine the size of the aerosol particle.

Functional principles

An aerosol spectrometer operates by leading single aerosol particles through a light beam or through an intensively lighted measuring volume. The light pulse which is scattered by the single particle is measured and also its intensity. Knowing the sample volume flow and the measuring period the particle number concentration can be derived from the number of the counted scattering pulses. The intensity of the scattered light can be interpreted to a particle size. Depending on calibration procedure and information about the micro physical properties of the measured aerosol particles it is also possible to calculate the particle mass for a given particle size fraction.

There are many types of aerosol spectrometers commercially available, differing in their technical setups and main application fields. In the following the main components of an aerosol spectrometer will be explained, with regards to different technical solutions.

As light source either lasers (e.g. diode-lasers, He-Ne-lasers) or intensive white light (e.g. xenon high-pressure lamps) is used. The light has to be focuses by a beam optic into a well defined, homogeneously illuminated optical measuring volume. The beam optic has to full fill several aims. Aerosol spectrometer using a laser have to compensate inhomogeneity of the laser beam intensity, caused by the Gauss-shaped distribution over the laser beam diameter. Also the border zone error, caused by aerosol particles which not fully pass the optical measuring volume or pass the laser beam in an area where its intensity is lower and the coincidence error, caused by two or more aerosol particles in the optical measuring volume at the same time have to be avoided (e.g. by an optical measuring volume limitation) or minimized. Beside the optical limitation the aerosol flow can be focused into the light beam, to achieve a so called aerodynamic measuring volume limitation.

After the optical measuring volume the beam might be collected by a light trap.

The aerosol flow passing the light beam in the measuring volume has to be strictly volume controlled, to assure the known time while an aerosol particle stays in the measuring volume and to be able to calculate the particle concentration in the sample.

The scattered light is collected by the detector optics under a certain solid angle with known aperture and led on a detector, typically a photodiode or a photo-multiplier. Aerosol spectrometers are available with detectors placed under different angles, namely in backward (appr. 0°-45°), forward (~180°) or 90° direction. The detector position and aperture can be chosen in a way to minimize the influence of refractive index on scattering intensity and to compensate Mie-interferences in scattering intensity when laser light is used. On the other hand signal to noise ratio of the scattering signal also is strongly influenced by the detector position and aperture angle. Finally the signal has to be amplified and analyzed by a pulse height analyzer.

The setup of an aerosol spectrometer will influence the specification such as precision, particle size range, number of size classes, maximum concentration range but also price, costs for calibration and service, size and weight or roughness. Beside the technical specification of the optical detection mentioned above, aerosol spectrometers are available with different setups for special applications, e.g.:

- automatic coincidence indication, to increase counting efficiency
- isokinetic sampling inlets for measuring in high velocities
- implementation of Nafion dryers to reduce humidity in sampling air without heating and without affecting particle concentration or size distribution

- integrated filters to validate particle mass calculation or enable further investigation of analysed aerosol particles (gravimetrically, microscopically or chemically)
- ex-proof or temperature resist sensors to enable measurements in critical atmospheres and conditions
- battery powered for portable, mobile use

If a device measures the scattering intensity not from single particles but from multiple particles this photometers or nephelometers are not able to determine particle size distribution. Also particle concentration only can be measured with these systems, when certain requirements are met (particle size distribution and refractive index must be constant during a series of runs). This is for many applications most unlikely, so this type of photometers must not be mixed with aerosol spectrometers.

Calibration

Actually there is no national or international standard for calibration of optical aerosol spectrometers, yet. But both international (ISO) and national (VDI) working groups have been initialised to close this gap. The aim of these guidelines is to describe a calibration procedure and a validation method of aerosol spectrometers, to minimise the deviation in the results measured by a single aerosol spectrometer and to minimize difference in the results measured by different instruments.

Particle size can be calibrated by mono disperse standard particles, poly disperse aerosols in combination with cyclones (Binning J. et al. 2006) and theoretically by calculating calibration curve (scattering intensity vs. particle size for given refractive index or materials). For the theoretical calculation it is fundamental to now all parameters of the optical setup, e.g. published by the manufacturer, otherwise the results are completely wrong as published e.g. by Vetter T. 2004.

Two different types of size standards so called primary and secondary standards are available: Mono disperse, spherical reference particles, certified by the manufacture as primary calibration standards mostly in aqueous suspension, like polystyrene-latex (Boundy R. H. and Boyer R. F. 1952). The size of these particles usually was examined microscopically (light or electron microscope). A second possibility to use standard particles is to generate mono disperse reference particles directly into the gas phase, a so-called secondary standard. The users are responsible on its own to full fill repeatability and accuracy of this method. The dispersion of an aerosol using a generator is influenced by the electrically charge of the particles, so the standard aerosol must be neutralised by suitable methods. Water coatings on the standard particles, stabilisation from the solution or agglomerates of standard particles might cause problems during calibration procedure (Haller P. 1999).

For the calibration of particle concentration no primary standard exists. The counting efficiency only can be determined by comparison of an aerosol spectrometer with a well-defined reference unit (aerosol spectrometer or other e.g. Differential Mobility Analyzer, DMA and Condensation Particle Counter CPC).

Application examples for particle counting, sizing and mass determination

The selected examples show results from particle measurements both counting, sizing and mass determination. All measurements have been carried out with laser aerosol spectrometers of various models manufactured by Grimm Aerosoltechnik, Ainring.

All Grimm aerosol spectrometers operate with a very similar optical detection principle:

- diode-laser with 780 nm or 683 nm
- 15 channel or 31 size channels
- 90° scattering light detection with a given aperture of about 60°
- maximum particle concentration without coincidence 2,000 particles/cm³
- 1.2 l/min sampling flow

The spectrometers are available as portable model measuring particle concentration or particle mass for all size channels and equipped with an integrated 47 mm-PTFE filter for gravimetrically control of dust mass. Other Version is 19" rack version for continuously measurements of PM10, PM2.5 and PM1 (simultaneously) or particle concentration in 31 size channels. These models are equipped with a special sampling pipe to avoid condensation and to control relative humidity and optionally with external sensors, special weather housing for continuously measurements.

Figure 1 and 2 show a measurement during 24 hours in a laying hen stable in summer (figure 1) and winter (figure 2) performed with a 15 size channel aerosol spectrometer Model 1.108, Grimm and a time resolution of 1 minute mean values. The size channels where summed up to one fraction smaller 1 µm and one fraction bigger 1 µm.

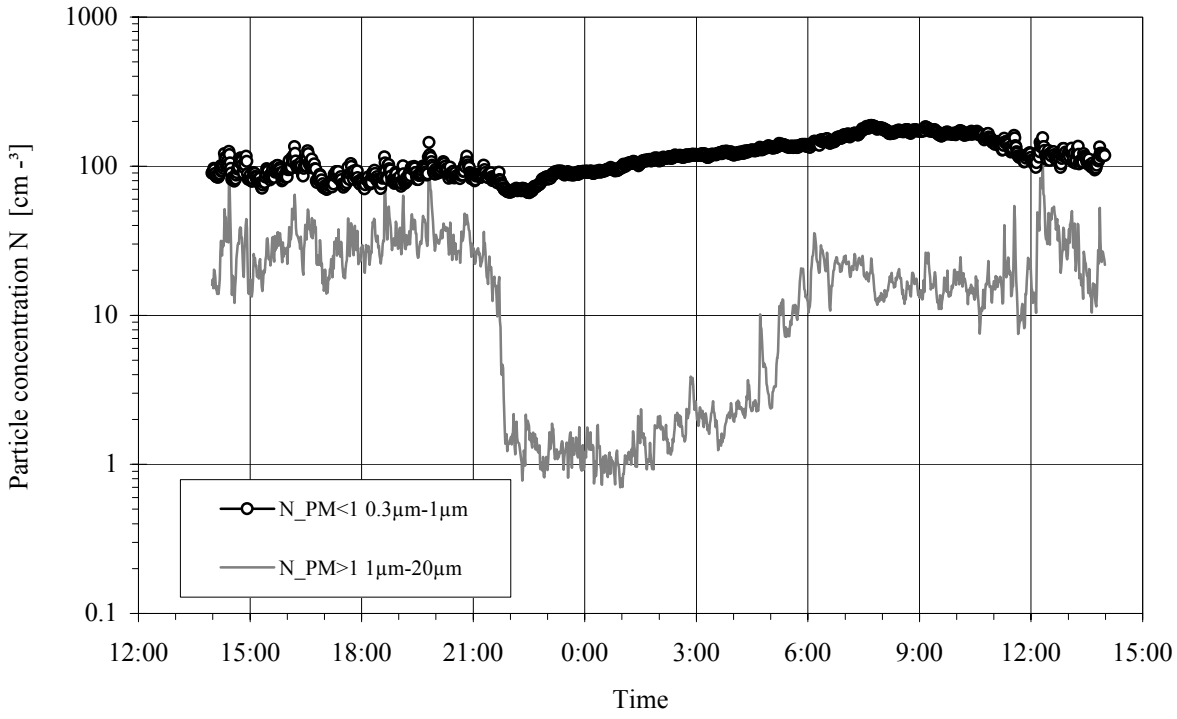

Figure 1:
Particle concentration in summer time in a laying hen stable for two size ranges (0.3 µm - 1 µm and 1 µm - 20 µm) during 24 hours, with a time resolution of 1 minute (Schneider C. 2005)

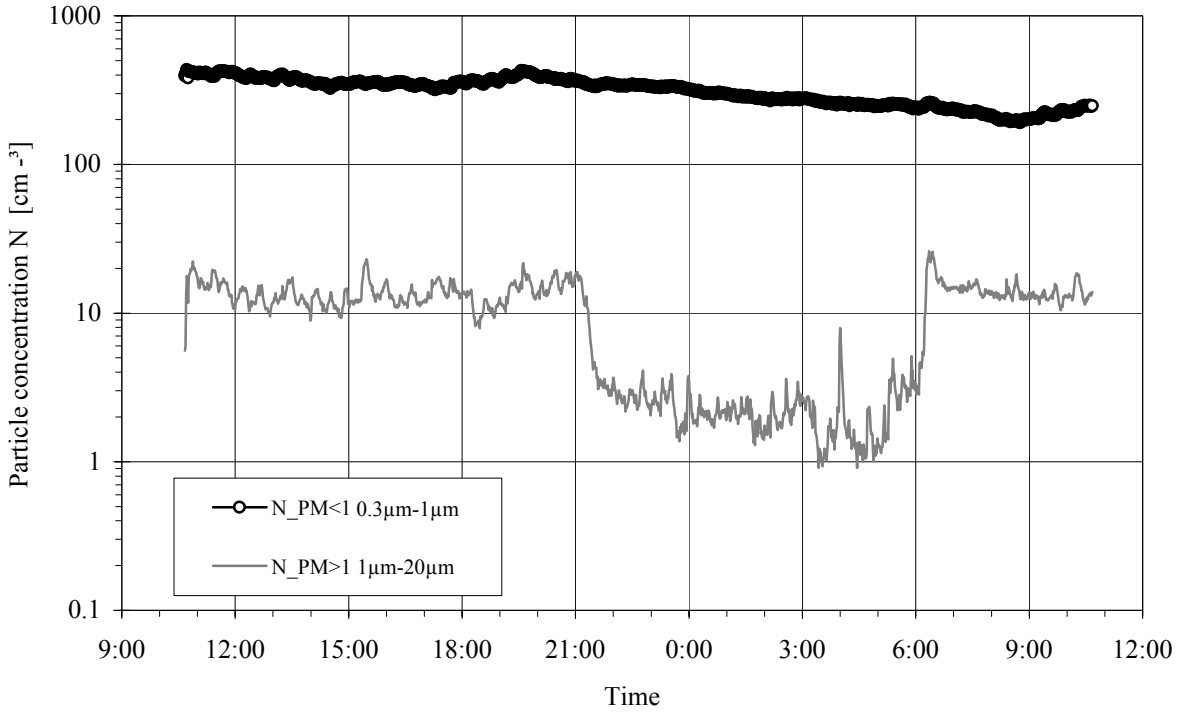

Figure 2:
Particle concentration in winter time in a laying hen stable for two size ranges (0,3 µm - 1 µm and 1 µm - 20 µm) during 24 hours, with a time resolution of 1 minute (Schneider C. 2005)

The figures above clearly show the differences in particle number concentration between the seasons. High ventilation rates during summer (figure 1) cause a dilution of particles smaller 1 µm. During night a seasonable influence on course particles can be neglected. Size selective concentration measurements with high temporal resolution containing a lot of information about air quality, ventilation performance or animal hygiene.

Figure 3 and 4 show a measured normalized particle size distribution with a Grimm Wide Range Aerosol Spectrometer consisting of an aerosol spectrometer (Grimm Model 1.108) in combination with a SMPS system for nano particle counting (CPC, Grimm Model 5.400) and sizing (DMA, Grimm Model 5.500). The measurement took place in a cattle stable (figure 3) and in a pig stable (figure 4).

The combination of optical particle counters with a SMPS system enables to measure a complete size range from 5nm up to 20 µm with one set up.

The graphs in figure 5 and 6 show data sets from a simultaneous measurement of two environmental dust monitors, Grimm, mode l 107, during a winter day. These aerosol spectrometers are able to obtain PM10, PM2.5 and PM1 mass concentration simultaneously. The spectrometers were mounted in a special weather housing for temperature and humidity control. One spectrometer (figure 6) was equipped with a heated sampling inlet, the other spectrometer (figure 5) was standard version with no temperature modification.

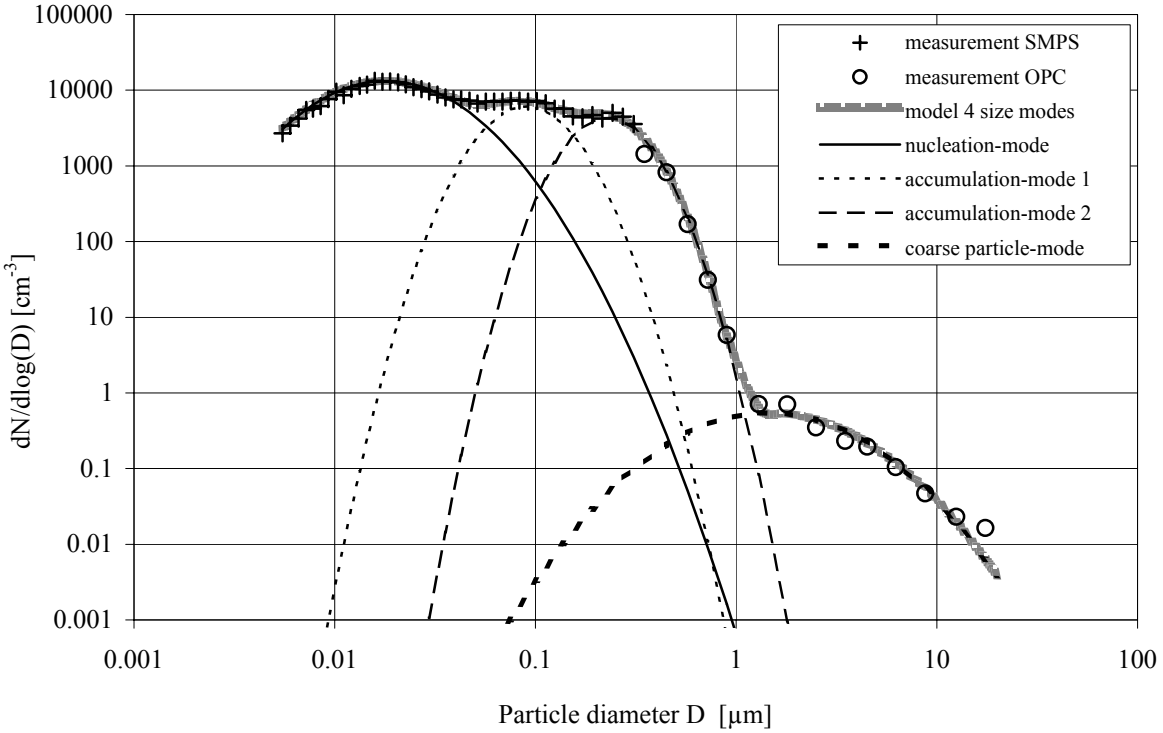

Figure 3:

Normalized particle size distribution in a cattle stable measured with an optical particle counter (OPC 0.3 µm < D < 20 µm) and a sequential mobility particle sizer (SMPS 0.0055 µm < D < 0.3 µm) and modelled particle size distribution (0.0055 < D < 20 µm) calculated for four lognormal size modes, namely nucleation-mode, accumulation-mode 1, accumulation-mode 2 and coarse particle-mode (Onyeneke-Edwards H. C. 2006)

The results in figure 3 and 4 show, that the optical particle counter and the SMPS system measure in the overlap size range around 0.3 µm in the same concentration range, also completely different techniques are compared (e.g. optical latex-equivalent diameter and electrical mobility diameter). This means that the used calibration methods for number and size are suitable and lead to precise and reproducible results. The measured size distribution easily can be explained by combined log normal distributions.

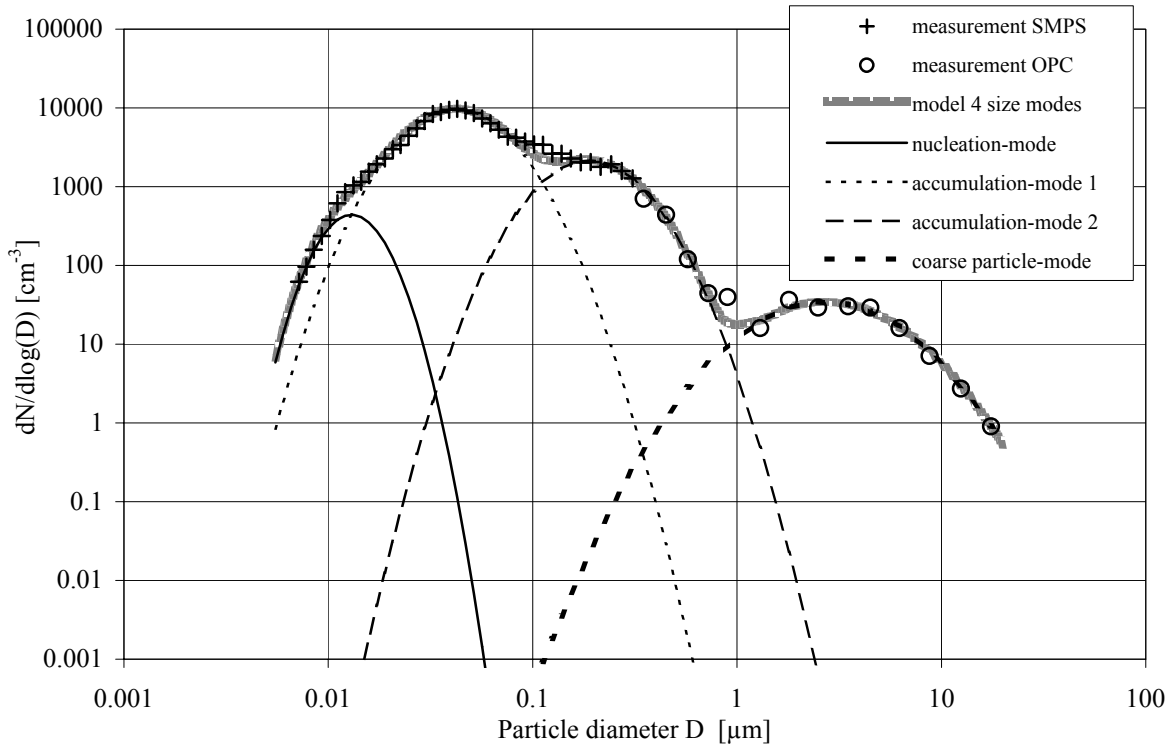

Figure 4:
Normalized particle size distribution in a swine stable measured with an optical particle counter (OPC 0.3 µm < D < 20 µm) and a sequential mobility particle sizer (SMPS 0.0055 µm < D < 0.3 µm) and modelled particle size distribution (0.0055 < D < 20 µm) calculated for four lognormal size modes, namely nucleation-mode, accumulation-mode 1, accumulation-mode 2 and coarse particle-mode (Mahmoud-Yasin N. 2006)

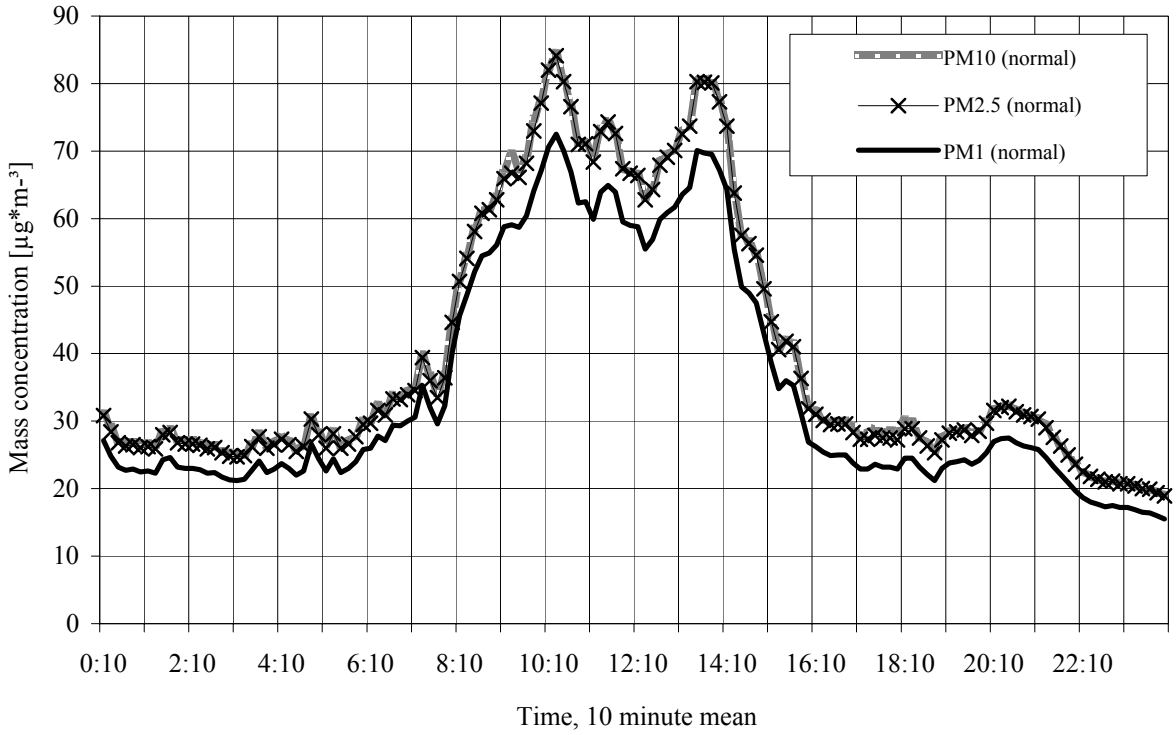

Figure 5:
Mass fraction PM10, PM2.5 and PM1 during a winter day, measured with a standard sampling inlet

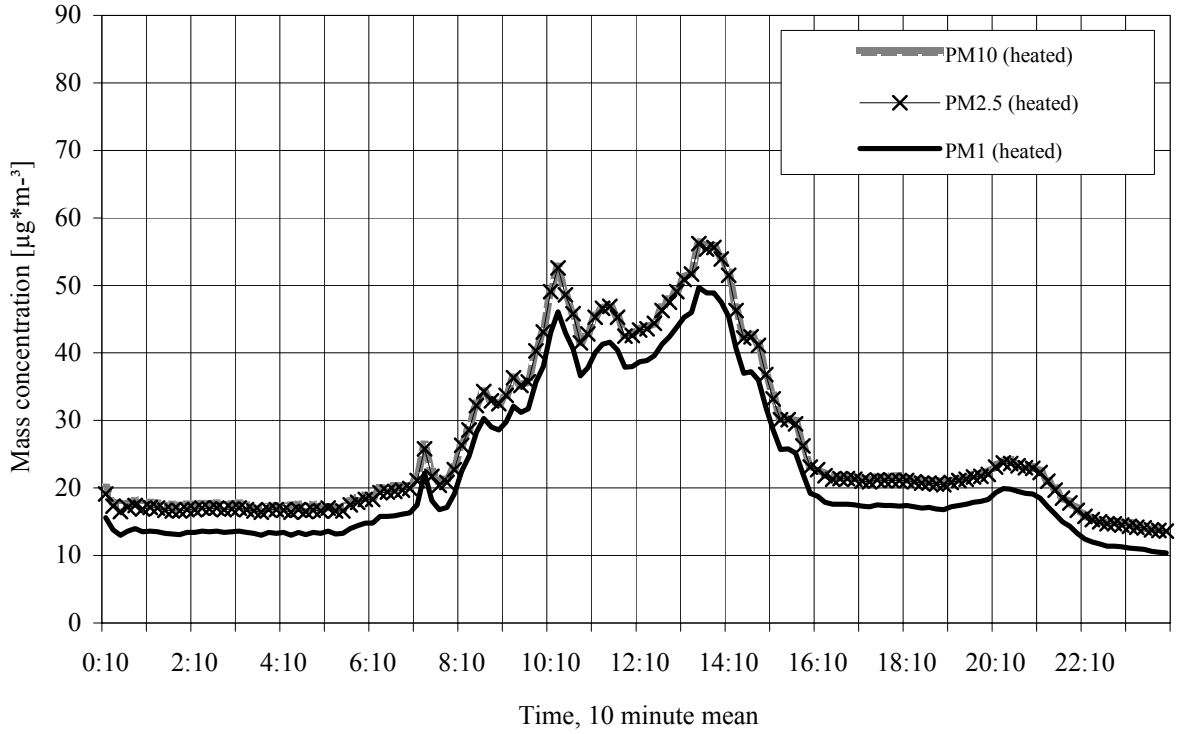

Figure 6:
Mass fraction PM10, PM2.5 and PM1 during a winter day, measured with a heated sampling inlet

As indicated in the graphs in figure 5 and 6 it can be seen, that PM10 and PM2.5 fraction are almost the same. Looking on the effect of the heated sampling inlet, it is obvious, that about 1/3 of the particulate matter is semi volatile compounds, which are lost, if the sampling inlet is heated. This measurements have been compared with mass fractions collected with standard filter samplers, running at the same location simultaneously and chemical analyses. The Grimm 107 was used in many international campaigns with very good results, e.g. Gorny R. L. et al. 2002, Liao C.-M. et al. 2003, Putaud J.-P. et al. 2003, Querol X. et al. 2004.

The further development of the model 107 is the Grimm model 180 environmental dust monitor. This stationary 19" rack version has been approved (Umweltbundesamt, 2006) as an equivalent PM10 measuring system, according to EN 12341! It runs with a included Nafion dryer, which enables simultaneously determination of PM10, PM2.5 and PM1 mass fraction, while the PM2.5 mass fraction includes the semi-volatile species, but excluding water. This is a big advantage in comparison to other environmental dust samplers as described e.g. by Delbert J. E. et al. 2007.

As shown above, aerosol spectrometers successfully can be used in applications, where particle number, size or mass has to be determained.

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Effects of different sampling heads such as PM1, PM2.5, PM10 and Sigma 2 on the particle size determination with aerosol spectrometers

L. Mölter[1], G. Lindenthal[2], and T. Hinz[3]

Abstract

From epidemiological research it is well-known that gravimetric emission and immission measurements do not supply all information which is crucial for the effects of atmospheric particles on human health. Counting measuring methods (e.g. optical aerosol spectrometers) offer the advantage of the particle size and particle quantity determination in time high resolution, thus quasi real time determination.

A representative sampling is crucial for the information content of the particle size and particle quantity measurement both during emission measurements (e.g. in exhaust ducts) and measurements in ambient air or in stables.

In exhaust ducts the particle sampling must be accomplished in an isokinetic and representative way concerning the particle size and particle quantity over the duct cross section. During the immission measurement with counting measuring methods the sampling concerning particle size and particle quantity has to be likewise representative. For the adherence to the appropriate definitions of particle fractions often pre-separators (e.g. sampling heads such as PM1, PM2.5, PM10) and passive collectors like Sigma 2 (VDI 2119 part 4) are used.

With the white-light-aerosol-spectrometer-system welas® both the representative sampling place in an exhaust duct and the collecting efficiency of different sampling heads can be found and/or determined fast and safely.

In this paper we are going to present results which were determined in the exhaust air duct of a turkey hen stable and in a turkey hen stable in November 2006.

Keywords: collecting efficiency, representative sampling, isokinetic sampling, pre-separator

1 Basics concerning isokinetic and representative sampling in exhaust air ducts

During the particle size and particle quantity determination of aerosols the selection of the measuring method, of the measuring device and the proceeding with the sampling are of crucial importance. The unclear answer to the following questions not seldom leads to serious errors during the particle size and particle quantity determination:

- What is to be measured?
- Where is to be measured?
- How is to be measured?

A typical measurement chain for the on-line measurement of particle sizes and particle quantities is represented in figure 1. This figure also shows that the weakest element of the measurement chain determines the quality of the measurement.

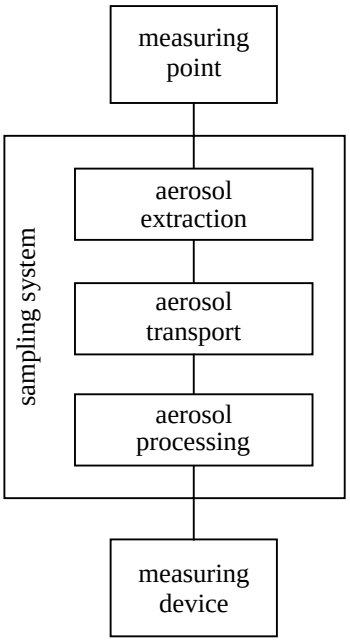

Figure 1:
Typical measurement chain

During the particle size and particle quantity determination in exhaust ducts (e.g. of forced ventilated stables) one must sample in an isokinetic and representative way. An

[1] Palas® GmbH, Karlsruhe, Germany

[2] Ingenieurbüro für Partikeltechnologie und Umweltmesstechnik, Espenau-Hohenkirchen, Germany

[3] Institut für Technologie und Biosystemtechnik der Bundesforschungsanstalt für Landwirtschaft, Braunschweig, Germany

isokinetic sampling makes sense only if the constancy of the flow rate at the sampling place is not higher than the indicated tolerance to the isokinetic sampling.

The effect of the non-isokinetic sampling can be easily made understandable with the help of figure 2.

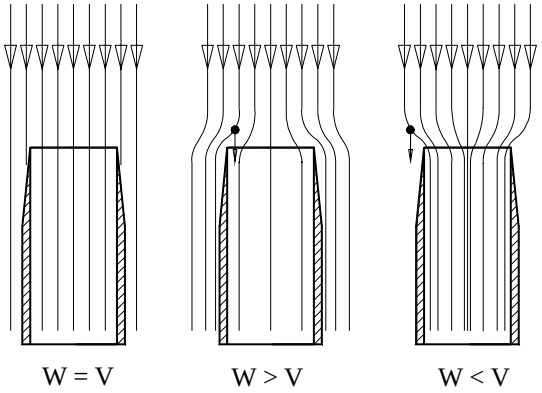

Figure 2:
Representation of the isokinetic sampling (W = flow rate in the duct, V = flow rate in the probe)

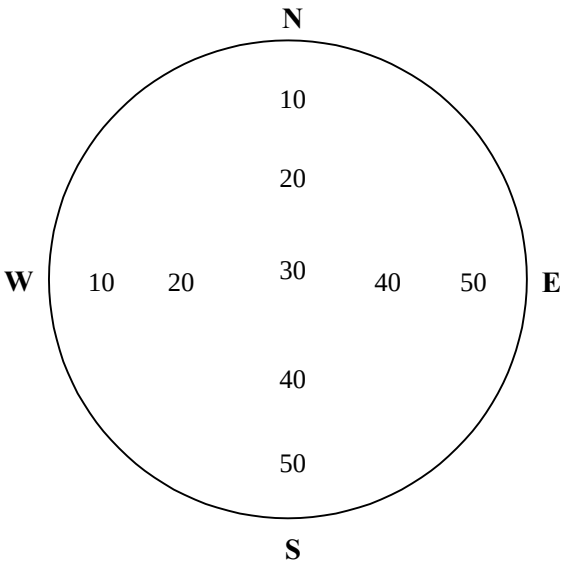

Figure 3:
Alignment of the measuring points in the exhaust duct, duct diameter 60 cm

Isokinetic case W = V: All particles, which flow against the probe within the flow cross-section, are collected. In case of a too slow suction W > V, more large (inertial) particles get into the probe opening. Those particles cannot any longer follow the flow lines due to their inertia. In case of a too fast suction W < V, more small particles are sucked in, since the large (inertial) particles not being able to follow the flow lines fly by at the outside.

The error of the non-isokinetic sampling is not linear and depends according to (Hinds W. C. 1999) on the ratio of the particle concentration C_V/C_W, on the ratio of the gas flow W/V and on the particle diameter (C_V = concentration in the sampling probe, C_W = concentration in the exhaust duct).

The representative sampling place concerning particle size and particle concentration can be determined fast with a counting measuring method at sufficient particle concentration by scanning the cross section.

Different particle losses in the sampling lines can occur by selection of the material (metal or plastic), of the inside diameter, of the length of sampling lines and by sedimentation and impaction effects.

1.1 Results for the determination of the representative sampling

The numbers in figure 3 represent the distance to the duct wall in cm from the north and/or west side. Thus, position 30 is the duct centre.

The incident-flow rate was measured with a thermo-anemometer. Since the incident-flow rate is not constant over the duct cross section, it was specified with 5 m/s for the setting of the diameter of the isokinetic sampling probe.

The particle size and particle quantity determinations were detected with:

optical aerosol spectrometer:	welas®-system 2000
	welas®-sensor 2300
sampling volume flow:	5 l/min
measuring ranges:	0.3 – 17 µm or
	0.6 – 40 µm
selected measuring range:	0.6 – 40 µm
measuring times:	at each position 2 x 60 s

The measuring range of 0.6 - 40 µm was selected on the one hand because particles as large as possible should be measured and on the other hand because the data should be compared with a parallel operated optical aerosol spectrometer of the company Grimm, whose measuring range was indicated from 0.5 to 30 µm.

With the welas®-sensor 2300 the particle concentration can be measured practically coincidence-free up to $C_N = 10^4$ particles/cm³. The measured particle numbers $\dot{N}$ increased from the outside inward from $\dot{N}$ = 1036 - 2423 particles/min.

Due to the particle number distribution in the exhaust duct, the sampling point for the welas®-system was defined in position 40 of the N-S axis and the suction flow rate for the gravimetric mass determination in position 20 of the W-E axis.

The Grimm-spectrometer was attached with an isokinetic part flow consumption probe in the suction flow rate for the

gravimetric mass determination. Over the measuring time of 15 min both the welas®-system and the Grimm-spectrometer could register at the same time the fluctuations of the particle numbers. Afterwards the welas®-system was attached at the part sampling probe of the Grimm-spectrometer, in order to determine there the particle size distribution. Following, an axial cyclone (Hinz T. 1983) with the separation function according to figure 4 was inserted in front of the part sampling probe, in order to determine its separation function.

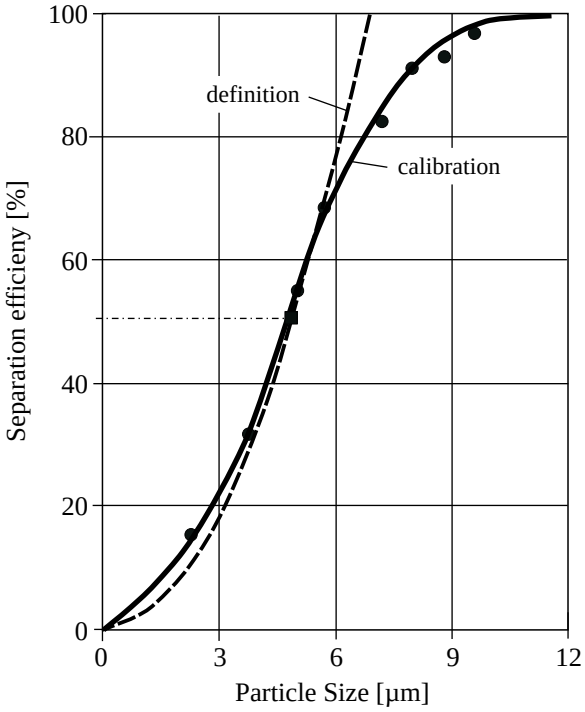

Figure 4:
Johannisburger separation function
Theoretical curve and actual calibration while sampling with a flow rate of 50 m³/h

For this cyclone the cut-off-diameter is indicated with 5 µm. All particles larger than 7.5 µm should be separated.

In figure 5 the measuring curves are represented which were measured with the welas®-system after the part flow consumption with and without measuring cyclone. Since the theoretical separation function (figure 4) of the cyclone does not let pass practically any particles above 10 µm, the measuring range at the welas® was selected from 0.3 - 17 µm.

Figure 5 shows clearly that the cyclone has a cut-off-diameter of 5 µm and does not let pass practically any particles larger than 10 µm.

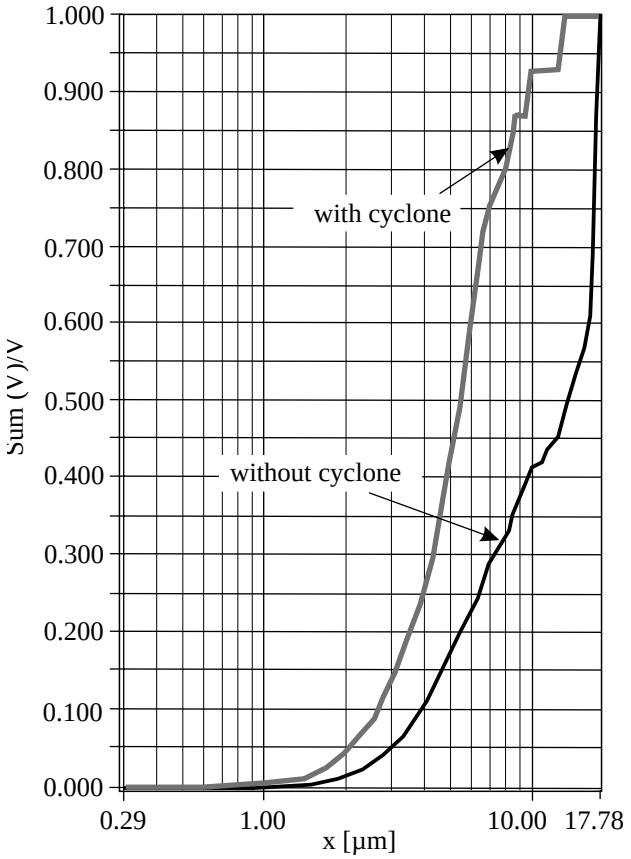

Figure 5:
Measured relative volume distributions of the particles in the exhaust duct, welas®-sensor 2300 behind the part sampling measuring section with and without cyclone, measuring range 0.3 – 17µm

2 Definitions concerning separation devices and separators

The evaluation of separation degrees and separation efficiency curves can be quite complicated on closer inspection, since here the dependence of the separation process on the particle size and possibly also still further material and operation properties are to be considered. The selected measuring method and the measurement setup can affect the separation degree and the separation efficiency curve likewise substantially.

The separation degree (figure 6) indicates which portion of the feed arrived after the separation in the coarse fraction and/or in the fine fraction.

The mass balance during the two-fraction separation, i.e. during the separation with a separation cut (1 coarse fraction and 1 fine fraction), is very helpful during the evaluation of separation devices and separators.

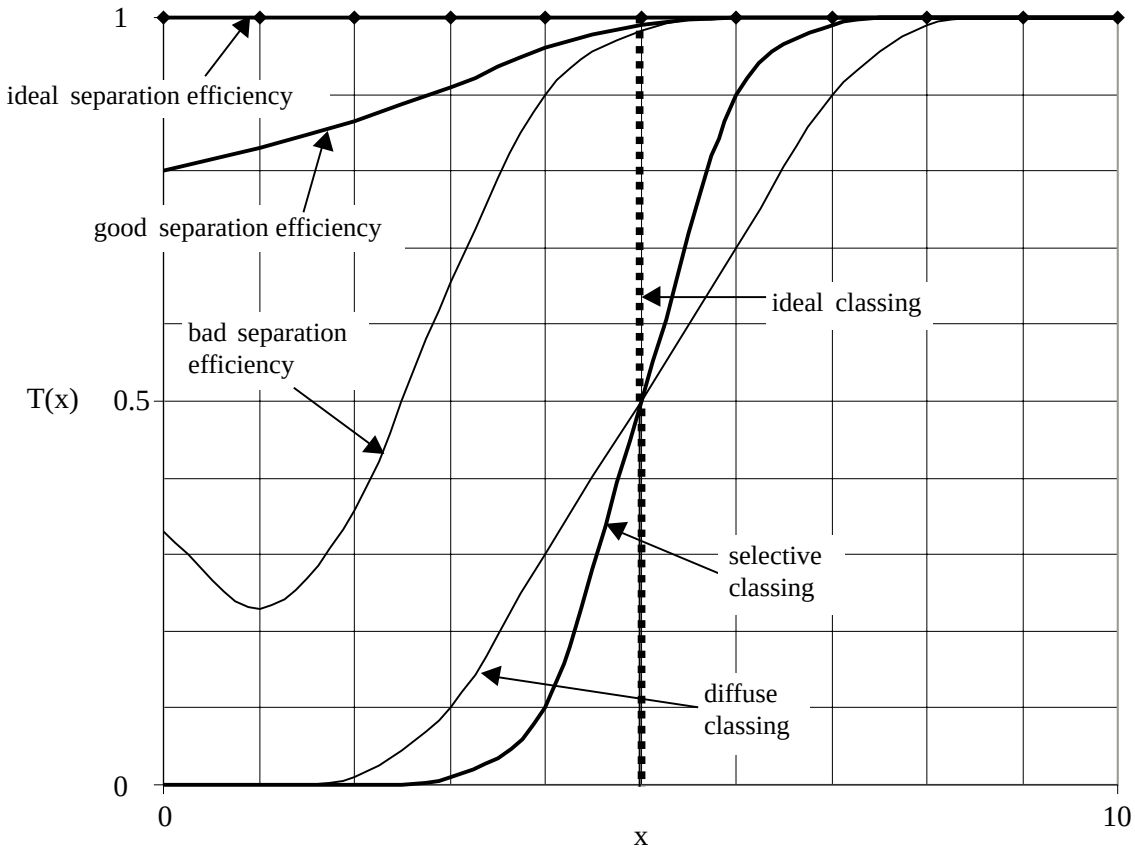

Figure 6:
Separation curves of separation devices (e.g. cyclones) and separators (e.g. filters)

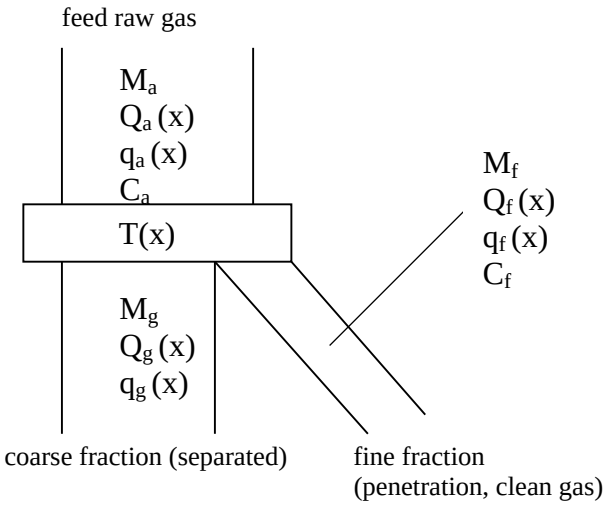

Figure 7:
Mass flows of a separation

The integral mass balance supplies: $M_a = M_a + M_f$

The total separation efficiency: T $\widehat{=}$ coarse mass fraction

$$T = \frac{M_g}{M_a} = \text{Retention}$$

The total penetration grade: P $\widehat{=}$ fine mass fraction

$$P = \frac{M_f}{M_a} = \text{Penetration}$$

If one refers all masses to M_a, then one gets: $1 = T + P$

With the dust concentration $C_{raw} = C_a$ in raw gas and $C_{clean} = C_f$ in clean gas one can also write:

$$T = 1 - \frac{C_{clean}}{C_{raw}}$$ T = total separation efficiency

$$T(x) = 1 - \frac{C_{clean}(x)}{C_{raw}(x)}$$

T(x) = separation degree depending on the particle size $\widehat{=}$ fractional separation efficiency

$C_{clean}(x)$ = particle concentration in clean gas depending on x

$C_{raw}(x)$ = particle concentration in raw gas depending on x
x = particle size

A classifier can be described meaningfully only with a separation curve. Just like during the description of a particle size distribution at least two parameters are needed: a location parameter and a distribution parameter.

Often, only the location parameter T(x) = 50 % is indicated as value of the separation limit, cut-off. Since separators frequently do not show a symmetrical run of the separation curve (figure 6), it has to be advised against this approach. The exclusive indication of the separation limit in point T(x) = 50 % does not supply any information about which form shows the separation curve and/or which selectivity shows the separation device.

The selectivity is described by the distribution parameter χ.

This parameter can be calculated at different particle sizes:

$$\chi = \frac{x_{25}}{x_{75}}, \quad \chi = \frac{x_{16}}{x_{84}} \quad \text{or} \quad \chi = \frac{x_{10}}{x_{90}}$$

E.g. x_{10} means the particle size x at which the separation curve is T(x) = 0.1 = 10 %.

The values x_{10} to x_{90} describe the run of the separation curve within a certain range below and/or above x_{50}. The more closely is selected this range, the fewer information contains the curve on the border areas of the separator.

For the critical evaluation of the selectivity and/or of the cut-off of a separation device or of a separation efficiency curve the meaningful indication of the used measuring technology and a clear sketch of the measurement setup are necessary. For this measuring task the welas®-system proved particularly of value, since here also large particles can be clearly detected.

A separation device has always a separation curve, and no step function, as this is described also in (John A. C. et al. 2001 and VDI 2066 part 10).

2.1 Measurement results related to collecting efficiency of sampling heads

With the welas®-system, one determined the different collecting efficiencies of the sampling heads and/or without sampling head but only with sampling tube with an inside diameter of 8 mm.

It was determined in a measurement-technological way that the particle number within the measuring range from 0.6 - 40 µm with the Sigma 2 according to VDI 2119 part 4 is around the factor 2 higher than with only the sampling tube of diameter 8 mm. After these measurements, the different sampling heads PM1, PM2.5 and PM10 were connected with the welas®-system and their collecting efficiencies were determined.

In figure 8 the respective relative cumulative frequency distributions of the measurements with the individual PM-heads are represented. The different separation functions are clearly recognisable.

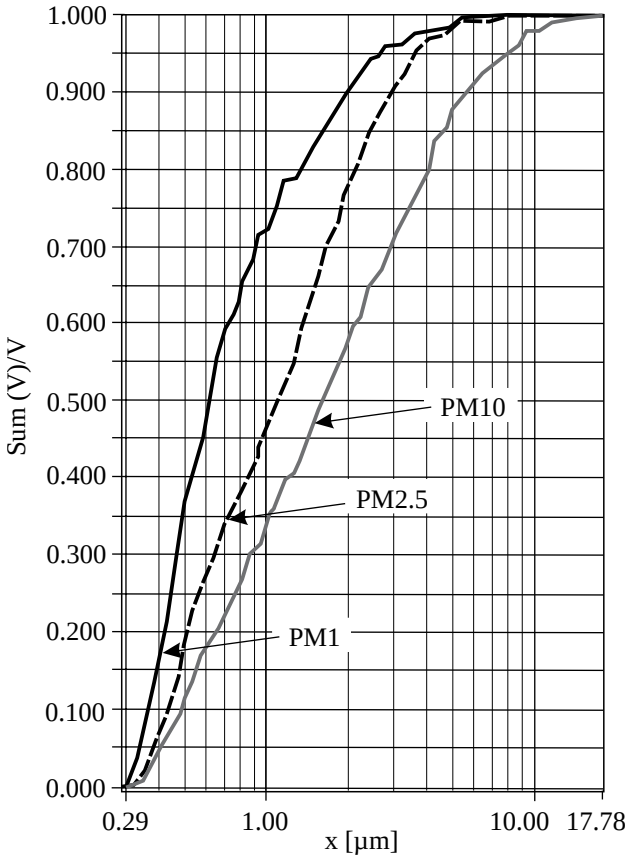

Figure 8:
Relative number cumulative distribution in the turkey hen stable with different PM-heads

Figure 9 shows the relative volume distributions of the particles let pass by the different PM-heads. One can clearly recognise that this is also a separation curve and not a step function.

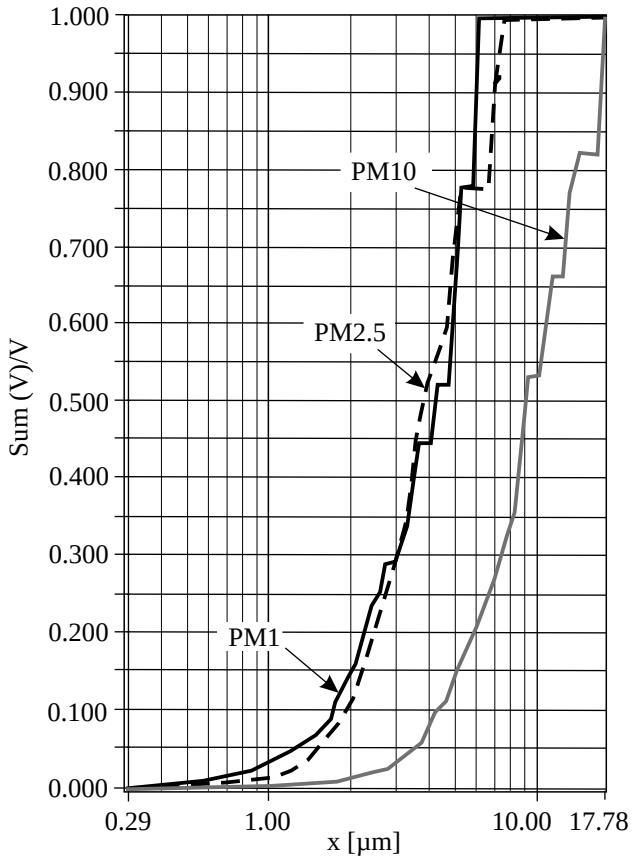

Figure 9:
Measured relative volume distributions in the turkey hen stable with different PM-heads

These measurement results confirm the definition and representation in VDI 2066 part 10.

PM10	$d_{50/3} \leq 10$
PM2.5	$d_{50/3} \leq 2.5$
PM1	$d_{50/3} \leq 1$

There is a variation of definition at the result of PM1.

Table 1:
$d_{50/3}$ values of the measured PM-heads

PM	10	2.5	1
$d_{50/3}$	9 µm	2.5 µm	2.5 µm

During the sampling with Sigma 2 there were measured a higher number of particles and larger particles than with the PM10-head. Thus, at the discussion on the evaluation of the total dust examination the sample collector Sigma 2 is to be absolutely considered.

For the evaluation of a measured separation function of a separator it is necessary to know the device characteristics of an aerosol spectrometer.

In the VDI 3867 part 4 draft and in the ISO/CD 21501-1 e.g. the size resolution, the size classification, the border zone error as well as their effect on the particle size distribution are described. These new guidelines will be useful for the users of particle measuring technology with counting measuring methods for the determination of the mass concentration.

3 Conclusions

With optical aerosol spectrometers the collecting efficiency difference of different sampling heads and the function of pre-separators can be determined fast. Also the representative sampling place in exhaust tubes can be found fast and safely.

The measurements shown above including setup and dismantling were accomplished in 6 hours.

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ISO/CD 21501-1: Determination of particle size distribution – Single particle light interaction methods – Part 1: Light-scattering aerosol spectrometer

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VDI 2119 part 4: Measurement of particulate precipitations – Microscopic differentiation and size fractionated determination of particle deposition on adhesive collection plates – Sigma-2 sampler

VDI 3867 part 4 draft: Measurement of particles in ambient air – Methods for characterizing test aerosols – Determination of the particle number concentration and particle size distribution – Optical aerosol spectrometer

Advantages and limits of aerosol spectrometers for the particle size and particle quantity determination stables and air exhaust ducts

L. Mölter[1] and M. Schmidt[1]

Abstract

For a reliable particle size and particle quantity determination in particle concentrations up to 10^5 p/cm³ optical aerosol spectrometers (OAS) offer many advantages. However, they must fulfil certain technical requirements in order to ensure significant measurement results. This essay imparts important basics for reliable particle measurement with optical aerosol spectrometers. The technical set-up and the function of an OAS are described and possible sources of error are also mentioned.

In order to avoid errors, a new OAS was developed whose measuring procedures are based on the "true" Mie-scattering, i. e. on the scattering at the spherical single particle. This measuring instrument offers clear calibration curves and very good device characteristics such as size resolution and classification accuracy, measures without border zone errors and has coincidence detection. The sensors can be used into temperatures of – 90° to 70°C. With a special cuvette, particles can be measured in overpressure up to 10 bar and in hot gases up to 120°C.

With this OAS an isothermal particle measurement is possible. Therefore, the system offers special advantages for the clear and economic characterisation of MDI, DPI, inhalers or jet mills as well as of separators even in highly explosive environments or in badly accessible places.

Keywords: high particle concentration, function OAS, explanation and influence of device parameters on particle size distribution, calibration curve, border zone error, coincidence

1 Introduction

At the particle size analysis, counting measuring methods are naturally used for numerous applications, because measurements at the particle collective or gravimetric procedures, which come from the total mass as valuation criterion, do not supply all desired information depending upon the application. Thus e.g. a photometer cannot differentiate between a particle size and a particle concentration variation. With counting measuring methods however, a time resolution of the particle sizes and the particle concentration and thus a clear determination of these measurable variables is possible.

For the determination of the fractional separation efficiency of separators, the size and the number of particles both in raw gas and in clean gas must be determined accurately, e.g. at blow-by in-situ measurements, at tests of interior filters or engine filters according to ISO 5011. Here the use of optical aerosol spectrometers (OAS) is recommended (Hess W. F. et al. 2005, Mölter L. et al. 2003, Baron P. et al. 2001, Hinds W. C. 1999, Hemmer G. et al. 1999, Umhauer H. et al. 1995), which can measure also in high particle concentrations up to 10^5 P/cm³ without dilution. OAS offer special advantages also for the characterisation of MDI, DPI and nebulisers in pharmacy, the production monitoring of active substances, the cut off determination of impactors, cyclones (Binnig J. et al. 2005), impingers or the measurement of atmospheric aerosols (e.g. bio aerosols in stables, compost, fog or tunnel), particularly for the measurement of droplet aerosols.

An aerosol spectrometer is not to be mixed up with a so-called "clean room counter". The latter is to determine the number of particles in clean rooms - thus in low concentrations - within a short time, whereby the particle size determination does not have to be accurate.

An aerosol spectrometer however is used if a highly concentrated aerosol is to be characterised concerning the particle size and the particle number as clearly as possible. At present an ISO standard for optical aerosol spectrometers (ISO/CD-21501-1) and for clean room counters (ISO/FDIS 21501-4) is worked out.

In this essay, we will describe the technical set-up and the function of an optical aerosol spectrometer and we will explain the effects of the device characteristics on the particle size and particle concentration determination.

[1] Palas® GmbH, Karlsruhe, Germany

The particle measurement in environments with varying particle size distributions and varying particle concentrations makes special demands on the used measuring method. Examples for this are outside air measurements or measurements at objects with strongly varying raw gas concentrations such as cooling agent oil separators, in-situ measurements of the car engine or measurements of the lung function (inhale/exhale). In addition, measurements in very high particle concentrations or in environments accessible with difficulty and/or explosive environments require the employment of a measuring method particularly suitable for this purpose. Another application would be the quality assurance of a jet mill during the production of active substances.

With an optical aerosol spectrometer, which can measure at two different measuring points quasi simultaneously, there are completely new possibilities for the applications specified above. The new modular particle measuring system welas® 3000 measures with two sensors, which are connected to only one light source and only one photomultiplier, and obtains that way particularly quick and accurate results.

Thus, during filter testing, one of the sensors can be used for example in raw gas and the other can be used quasi simultaneously for clean gas measurement. Furthermore, unwanted factors, e.g. the influence of temperature and humidity on the particle size distribution, can be minimised with this measuring instrument. Due to the flexible selection of the sensors concerning their measuring volumes, this measuring system can be used for different particle concentration ranges. That way, it can be adapted optimally to the respective application.

2 Selection criteria for an appropriate measuring method

In particle measurement, counting procedures are suitable in particular if – from the beginning on – there are small sample quantities to be analysed, if there are not to high particle concentrations (number concentrations of less than 10^6 P/cm³) or if one has to measure in very small concentrations.

Counting aerosol measuring instruments are e.g.:

- OAS (Optical Aerosol Spectrometer),
- PDA (Phase Doppler Anemometer),
- Relaxation Time Spectrometer (Time of Flight Spectrometer),
- SMPS (Scanning Mobility Particle Sizer).

For the correct selection of the appropriate measuring method it is helpful to first visualise the requirements of the application to the measuring instrument to be used.

One has to expect e.g. at outside air measurements with a particle diameter of 0.2 µm a concentration of approx. 2000 particles per cm³. In such concentrations, the particle distance is clearly smaller than 1 mm (table 1).

Table 1:
Distances of the particles as a function of the concentration

Number N [m^{-3}]	Number N [cm^{-3}]	particle distance [cm]	particle distance [mm]	particle distance [µm]
1	10^{-6}	100	1000	
10^3	10^{-3}	10	100	
10^6	1	1	10	
10^9	10^3		1	1000
10^{12}	10^6		0,1	100
10^{15}	10^9		0,01	10
10^{18}	10^{12}		0,001	1

From this, one can conclude that e.g. an optical aerosol spectrometer with an optical measuring volume of approx. 1 mm³ cannot determine concentrations above 1 000 p/cm³ without coincidence errors, since with a particle distance of less than 1 mm there has to be allways more than one particle in the measuring volume.

By calculation, it can be observed that the particle distance in a concentration of 10^6 particles per cm³ amounts to 100 µm (table 1). Thus, if a particle concentration of 10^5 particles per cm³ is to be measured, then the border length of the optical measuring volume should be naturally not larger than 100 µm. The indication of the measuring volume size is very helpful for the user in order to be able to estimate in which maximum concentrations he can measure coincidence-freely. These examples show how important it is to know the theoretical basics for the selection of the appropriate counting procedure.

3 Theoretical basics of optical aerosol spectrometers

3.1 Lorentz-Mie theory

The measuring method of optical aerosol spectrometers is based on the Lorentz-Mie theory. The particle characteristic, the diameter, is determined by the impulse height analysis of the scattered light at the spherical single particle. The particle number is determined at the same time by the number of scattered light impulses.

Does light with the wavelength λ meet on a spherical particle with the diameter x and the refractive index m, then the light is scattered in different directions (figure 1).

The scattering of light at the particle is caused by diffraction, refraction and reflection. The polarisation plane of the incident light wave is also turned.

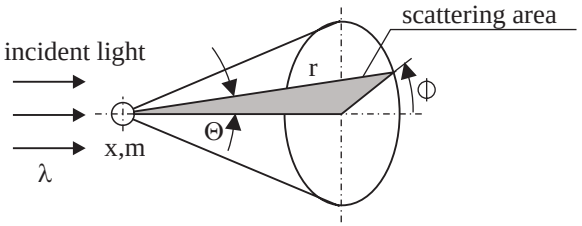

Figure 1:
Principle of incident light scattering

The intensity I of the light scattered at the single particles depends on the incident light intensity I_0, the polarisation angle Φ, the detection angle of the scattered light Θ, the refractive index n, the light wave length λ and the particle diameter x.

$$I = I_0 \cdot f(\Phi, \Theta, n, \lambda, x)$$

By means of the scattering parameter introduced by Mie (Mie G. 1908)

$$\alpha = \frac{\pi \cdot x}{\lambda}$$

the relation between the sphere circumference $\pi \cdot x$ to the wave length λ is used into the above mentioned equation:

$$I = I_0 \cdot f(\Phi, \Theta, n, \alpha)$$

With regard to the particle-size-depending scattering power one can differ between three ranges due to the introduction of the scattering parameter α:

a) Rayleigh-range: $\alpha << 1$; here the scattering power rises with the sixth power of the particle diameter and the scattered light will be proportional to d^6/λ^4. This means: If in the Raleigh-range one should be able to measure a half so large particle as before (lower detection limit), then the double supplied quantity of light

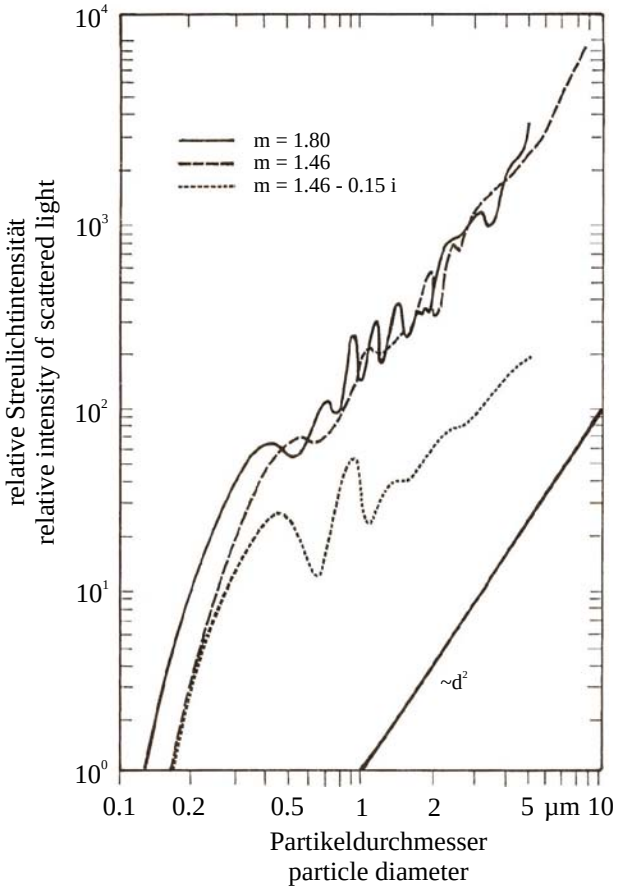

Figure 2:
Relative scattered light intensity for monochromatic light, e.g. laser light;
mean scattering angle θ0 = 45°; receiver aperture υ = 14;
light wave length λ = 0,436 µm; refractive index m
Source: VDI 3489 [13]

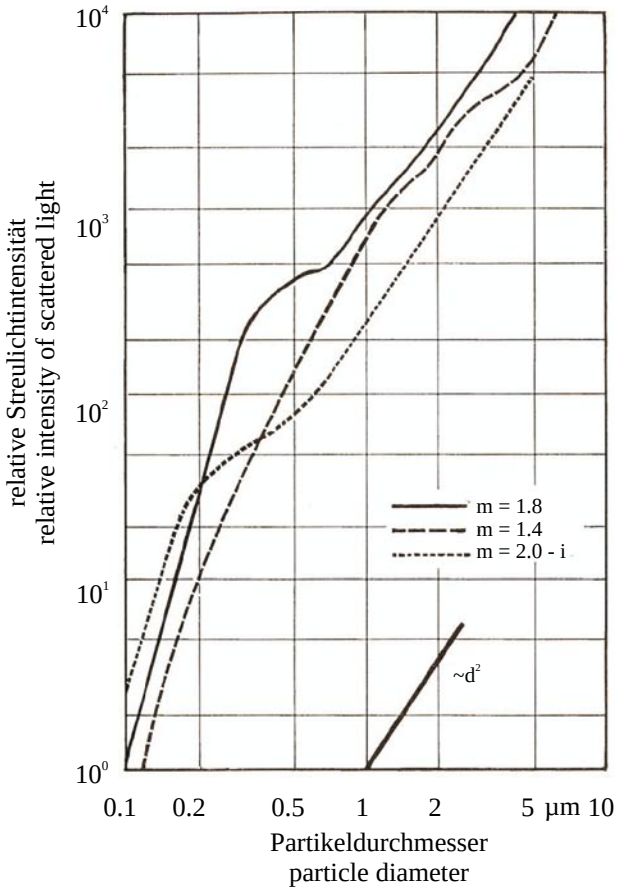

Figure 3:
Relative scattered light intensity for white light and 90° scattering;
mean scattering angle θ0 = 90°
receiver aperture υ = 24°
Source: VDI 3489 [13]

is not enough. The necessary quantity of light must be possibly 64-times stronger than at a twice as large particle.

b) **Mie-range**: $0.1 \leq \alpha \leq 10$; here the correlation between the scattered light intensity and the particle size is not clear (figure. 2). Whereas with white light and 90° scattered light detection this correlation is clear (figure 3).

c) **Fraunhofer- resp. geometrical range**: $\alpha >> 1$ (from $\alpha \approx 10$); here the quadratic correlation between the scattering power and particle diameter is valid.

In order to be able to determine with an aerosol spectrometer the particle size distribution as accurately as possible, a clear calibration curve (figure 3) is an important condition. OAS are usually calibrated by the manufacturer with monodisperse latex-aerosols with a refractive index of m = 1.59. Further suitable calibration procedures, e.g. the aerodynamic calibration, are described by Friehmelt (Friehmelt R. 1999).

3.2 Technical set-up of optical aerosol spectrometers

For the better understanding of the measuring method including the device characteristics, the set-up in principle of an OAS in forward scattering is represented in figure 4.

During forward scattering the light scattered by particles (figure 1) toward 180° is collected by the light source with a light-sensitive detector, e.g. a photomultiplier. At the 90° scattered light detection, the photomultiplier is attached orthogonally to the image plane. The height of the scattered light impulse is a measure for the particle diameter, while the number of impulses supplies the information on the concentration since the volume flow is known.

With the help of a lens system the light is focused on the desired measuring volume size. Before the receiver optics there must be installed a light collector in forward scattering, which protects the light detector against direct irradiation. Due to diffraction actions of the light and of the scattered light, this light collector leads to an ambiguous calibration curve, also when using white light. However, a source of white light in connection with a 90° scattered light detection secures a clear calibration curve for many refractive indices.

An advantage of a small measuring volume defined and projected with white light is that this one - in contrast to the laser beam - is homogeneously illuminated over the cross section. In the figure 5 and 6, real scattered light signals are represented from which the particle size is determined by detection of the impulse height. It is easily conceivable that the signal from figure 6 can be detected more easily and reliably than the signal in figure 5. Friehmelt points out in his dissertation (Friehmelt R. 1999; see also Friehmelt R. et al., 1998) that the particle size of irregularly shaped particles is determined with white light better than with laser light.

For the quality of the evaluated scattered light impulses, not only the optical set-up is responsible, but also the quality of the opto-electronic elements.

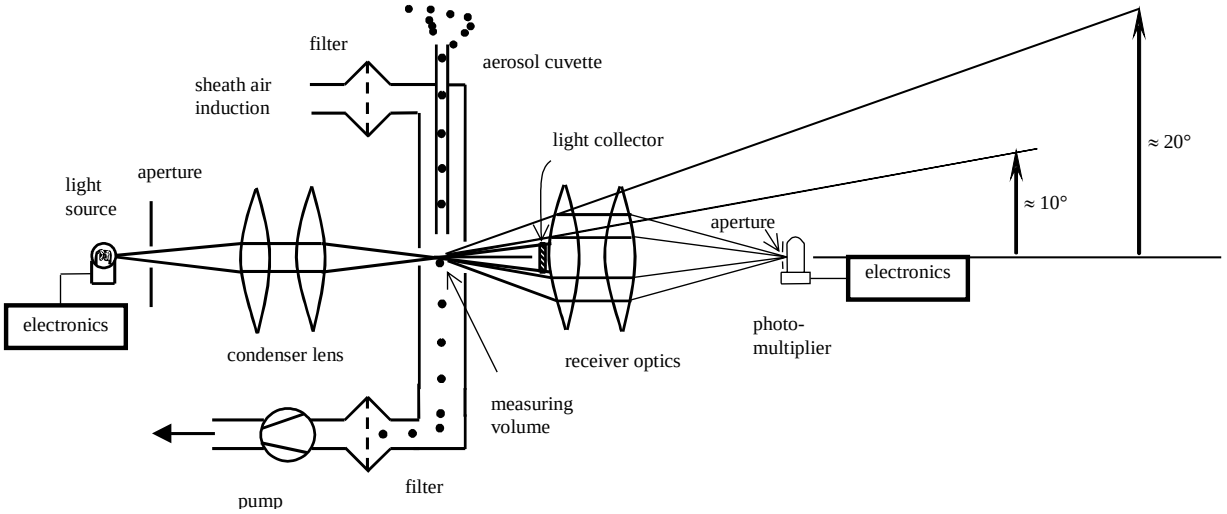

Figure 4:
Optical aerosol spectrometer (forward scattering)

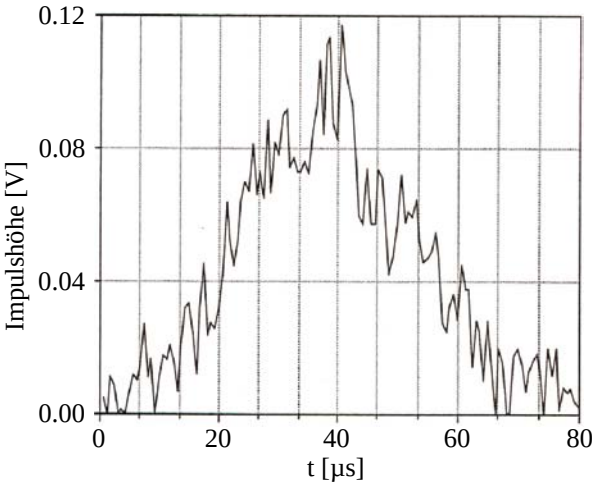

Figure 5:
Real scattered light signal at laser measuring devices: Signal of a 0,5 µm SiO2-sphere (monosphere, company Merck);
Source: H. Mühlenweg (Diss.)

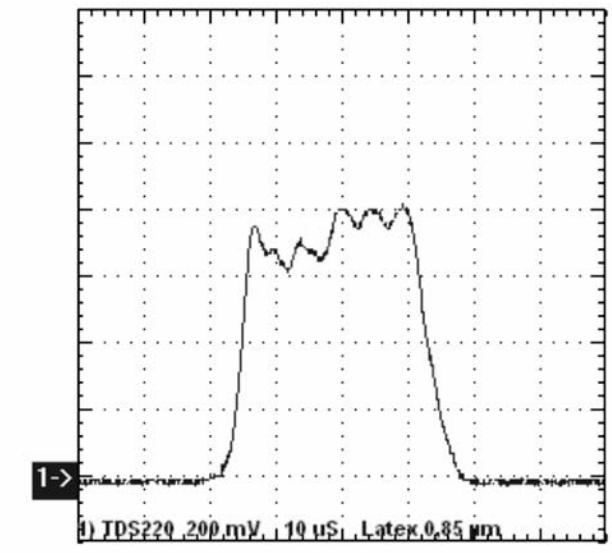

Figure 6:
Real scattered light signal at an homogeneously illuminated T-shaped measuring volume (white light source and 90° scattered light detection);
Source: Palas® GmbH

4 About the measuring accuracy of counting measuring methods

Independently of which counting method is used, usually one or more physical values must be known for the clear measurement of the particle size. Depending upon the counting method, these values are the following:

- the refractive index,
- the particle material density,
- the surface texture or the chemical composition,
- the electrical charge.

A particle measurement without the taking into consideration of the necessary physical values leads to different measuring results at different measuring methods. If different measuring instruments with the same measuring method supply different measuring results under the same test conditions, even when knowing the concerned physical values, then the cause might have to be found in the detail set-up and/or in the selection of the components of these measuring instruments. The shape influence of the particles plays a large role, too.

As generally in the measurement technique, it is meaningful also in the particle measuring technique to give information on the measurement uncertainty of the used measuring method and/or the concerned measuring instrument.

In the on-line particle measuring technique - in contrast to the mechanical measuring technique - it is practically impossible to determine measuring errors exactly with the help of a fixed tolerance (in the mechanical measuring technique for example the hole diameter of a drilling H7). Therefore, in the particle measuring technique, one has defined for the error consideration important device characteristics, which are specified in the VDI guide line 3489 and the ISO 13323-1 and will be described in the future in the VDI 3867 and the ISO/DIS 21501-1-4, too.

For a clear quantitative comparison of measurement results, which are won with different measuring methods, the used measuring instruments should be calibrated - if possible - with the same calibration procedure.

4.1 Particle size resolution and particle size classification accuracy

The particle size resolution gives information about which particle sizes can be differentiated from each other. The particle size classification accuracy indicates how exactly the particles are determined concerning their size.

4.2 Sources of error when measuring with optical aerosol spectrometers

Optical measuring volume limitations are used in practice in order to be independent of the Gauss intensity distribution of the laser light or to be able to determine as coincidence-free as possible the particle size distribution in high concentrations. The following possible sources of error must be considered:

4.2.1 Border zone error

Figure 7 shows the detail set-up of the measuring volume of an OAS with laser light and without optical measuring volume limitation.

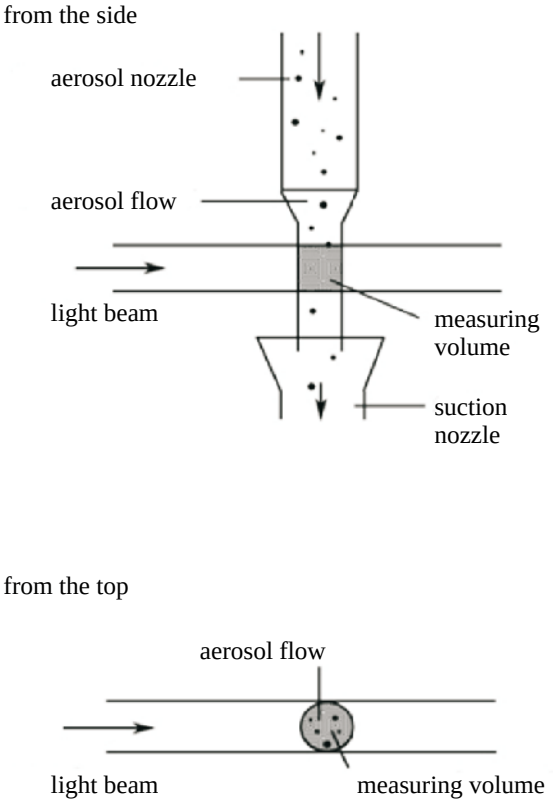

Figure 7:
Measuring principle: laser beam

The light intensity of a laser light beam is distributed Gauss-shaped over the beam diameter (figure 8).

A particle at the border of the laser beam scatters substantially fewer light than an equally large particle in the center of the laser beam. This so-called border zone error has been already known since 1970 and is described in (Helsper C. 1981, Blattner J. 1995, Mölter L. et al. 1995). This error can be minimised also by an aerodynamic focussing of the aerosol beam.

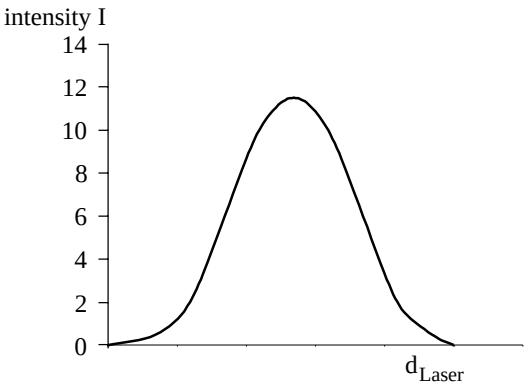

Figure 8:
Schematic representation of the intensity distribution of the laser beam (Gauss-shape)

If an optical measuring volume limitation is selected in order to minimise e.g. the effect of the Gauss distribution of a laser beam, then a further border zone problem arises. In figure 9 an optical measuring volume limitation is represented at which the problem of the border zone error can be made clear. A particle which is lighted only to 50% in the border zone scatters only half of the light of an equally large particle which is in the measuring volume center. Thus, the border zone error leads to the fact that particles are measured too small. In filter testing this leads to the fact that the separation efficiency is measured better than it really is because the fine parts of the particle size spectrum in the raw gas are measured too high.

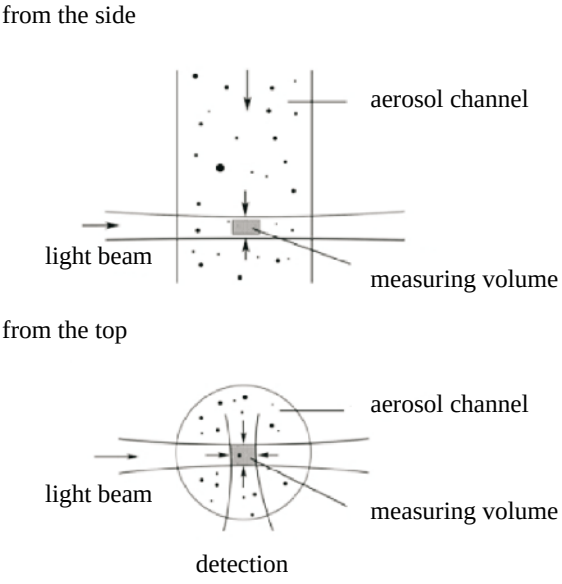

Figure 9:
Measuring principle: optical volume limitation, 90° scattered light detection

Figure 10 shows through the example of monodisperse test aerosols that the size of the border zone error is dependent on the particle size. The wider is the particle size distribution to be measured, the larger is also the border zone error.

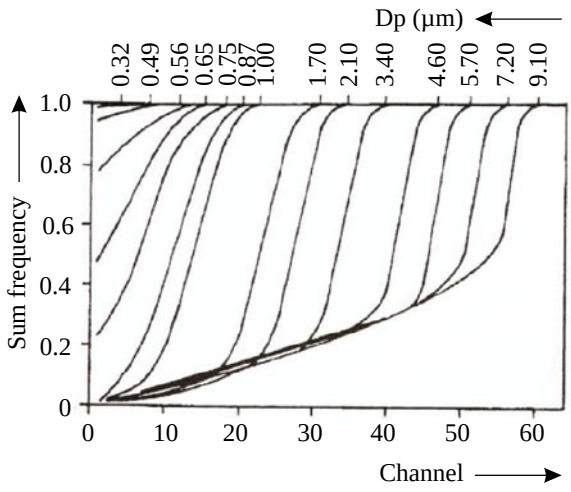

Figure 10:
Border zone error at optical measuring volume limitation (sum distribution of different mono-disperse aerosols)
Source: Helsper 1981 (Diss.)

4.2.2 Coincidence

One speaks about coincidence error if there is more than one particle at the same time in the measuring volume. If e.g. a laser beam diameter amounts to 1 mm, then one can measure with it maximally in a concentration of 1 000 particles per cm³ with negligible coincidence error, since, theoretically, the particle distance amounts here to 1 mm (table 1). Several particles in the measuring volume supply a higher scattered light impulse. Therefore the coincidence-afflicted particle measurement leads to two errors: The particles are measured too large and the concentration too low (Raasch J. et al. 1984, Szymanski W. W. 1996, Mölter L. 1995). At the conversion of the number distribution into the volume distribution these errors affect particularly strongly. In filter testing this leads to the fact that the separation efficiency is measured worse than it really is.

Thus, measurements without border zone error and with coincidence detection are an important precondition for an accurate and reliable filter testing.

5 Particle measurement without border zone error and with coincidence detection

The new **white-light-aerosol-spectrometer system welas®** has a clear calibration curve due to the white light source and the 90° scattered light detection (figure 3). Six different calibration curves for different refractive indices can be supplied with the welas® if desired. With the optical set-up of the system and its patented T-shaped measuring volume, with which it can be measured practically without border zone errors, a clear particle size determination with very good size resolution and very good classification accuracy is ensured (Lindenthal G. et al. 1998, Keusen G. 2003).

The measuring chamber is optionally heatable in order to avoid cross sensitivities and changes of particle size as a function of the relative humidity – an important point, because by a change of the relative humidity from approx. 40 % to 90 % e.g. hydrophilic particles such as NaCl particles can increase to around the factor 2 (Ebert E. et. al. 2002).

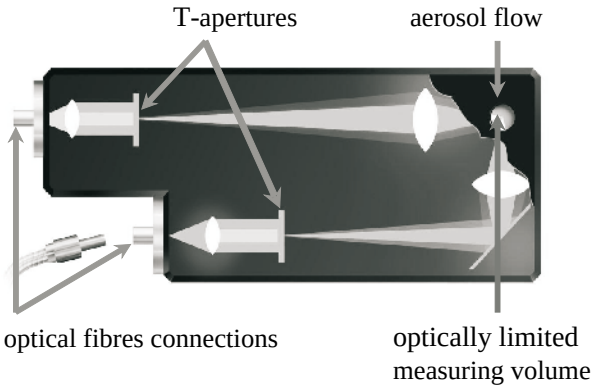

Figure 11:
Cross section through the welas® sensor

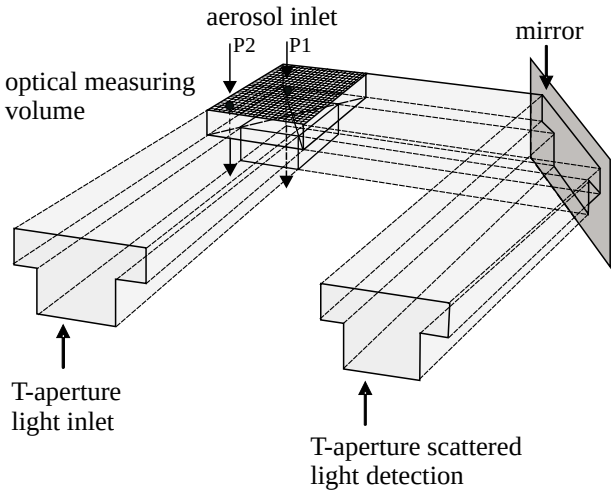

Figure 12:
Three-dimensional T-shaped measuring volume

In figure 11 one can see the compact set-up of the welas®-sensor. White light is injected over optical fibres into the aerosol sensor and focused with a T-aperture in a T-shaped way in the measuring volume. The scattered light is ob-

served over a T-aperture in an angle of 90° to the incident light. One receives a three-dimensional T-shaped measuring volume by the T-shaped light beam and the T-shaped observation level (figure 12). Since the aerosol is sucked with a vacuum pump through the sensor, one knows the runtime between inflow and outflow of the particles into the measuring volume.

5.0.1 No border zone error due to T-aperture technology

Figure 13 shows different particle positions in the T-shaped measuring volume as well as the corresponding scattered light impulse.

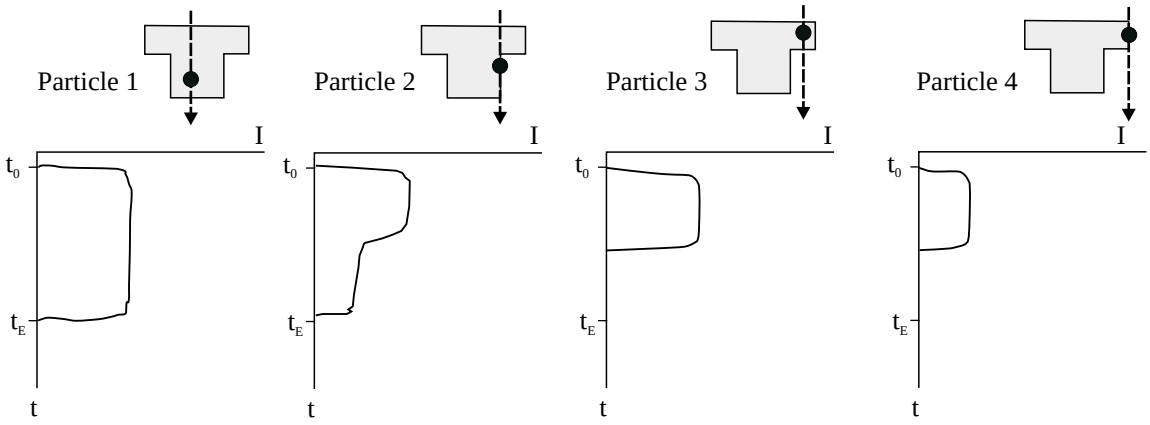

Figure 13:
Border zone error correction of the white light aerosol spectrometer welas®

Particles, which pass both the upper and the lower part of the measuring volume in a central place, generate a rectangular scattered light impulse as represented in case 1 of figure 13.

Case 2 represents a particle, which is still completely seized in the upper, larger part of the measuring volume, but is subject to a border zone error in the lower part of the measuring volume. The corresponding scattered light impulse shows a step-decrease with the passage of the particle from the upper to the lower part of the measuring volume. This particle is included with the higher impulse height into the evaluation.

In case 3 a particle is illuminated and seized measurement-technologically only in the upper part of the measuring volume. The signal evaluation recognizes this by the shorter impulse duration and rejects this impulse.

In case 4 a particle at the border of the upper part of the measuring volume is illuminated only to 50%. In this case the particle is measured around 50% too small. The measuring instrument rejects the signal of case 4, since the running time is too short.

The pulse width analysis is used beyond that also for coincidence detection.

5.0.2 Coincidence signal by means of T-aperture technology

If several particles fly at the same time through the measuring volume (figure 14), then one can easily imagine that the impulse height is higher than at only one particle of comparable size. Without coincidence detection the particles would be measured too large and the concentration too low.

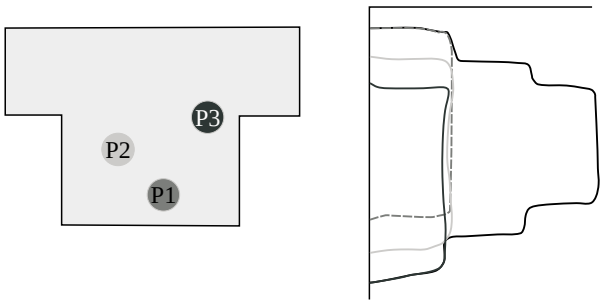

Figure 14:
Coincidence detection: particle flow through the optical T measuring volume and evaluation of the signal height and signal length

At the welas®-system (figure 14) the coincidence error is detected by the runtime measurement (the impulse is too long) and indicated by an acoustical and/or optical warning signal. Only a coincidence free particle measurement allows a reliable particle size and particle number determination of the aerosol. The user is responsible to use in accordance with the measuring task a particle measuring

instrument with the correct measuring volume size in order to measure as coincidence-free as possible. Due to the patented modular set-up of the welas®-system one can select depending upon measuring problem the optimal sensor with the optimal measuring volume size (border length 100, 200 or 300 µm) concerning the particle concentration to be measured (table 2).

Table 2:
Particle size range and maximum concentration range of the welas®-systems

	welas® 1100	welas® 1200	welas® 2100	welas® 2200
measuring ranges [µm]	0.18 – 40	0.18 – 40	0.3 – 40	0.3 – 40
concentration				
Cmax [p/cm³]	10^5	10^4	10^5	10^4

5.1 Evaluation of the scattered light signals

The evaluation of the scattered light impulses is done with the fundamental formulas of the error calculation and/or of the statistics known in the technology. The scattered light generated by the single particles supplies at the same time information about the number (single particle analysis) and the size of the particles, whereby the height of the scattered light impulse is a measure for the diameter and the number of impulses is a measure for the quantity of particles.

The better are the classification accuracy and the resolution of a particle measuring instrument, the narrower can be selected the characteristic category. Therefore, at the welas®-system the characteristic categories can be selected very closely. In principle, 4096 size classes are available for evaluation. The size classes are summarised into 32 channels per decade. The user can have the size distributions indicated in 32, 16, 8 or 4 size classes at the PC.

Due to this high resolution, with the welas® 1100 even tri-modal particle size distributions of highly concentrated droplet aerosols d_p < 2 µm can be measured as represented in figure 15. Here, this concerns the oil mist of a cooling agent lubricant.

For the representation of the particle size distribution neither complicated mathematics nor intransparent algorithms are used. The particles are assigned directly to the size classes (figure 15). Thus, e.g. the cumulative distribution is calculated according to the following formula:

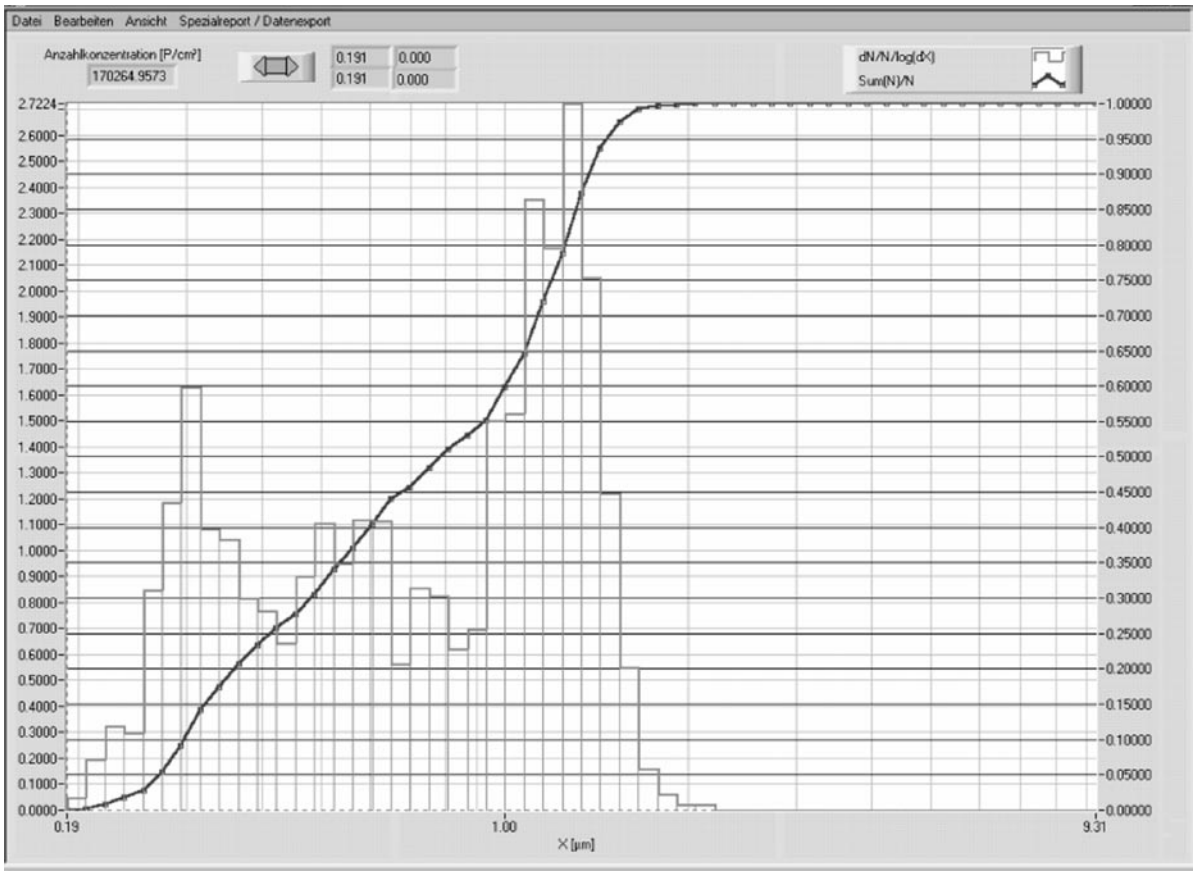

Figure 15:
Tri-modal particle size distribution

Cumulative distribution $Q_r(x)$

$$Q_r(x) = \frac{\text{quantity q of measured particles} \leq x}{\text{total quantity of particles } q_{ges}}$$

$$Q_r(x_i) = \frac{\sum_{i=1}^{n} q_i}{q_{ges}}$$

x = particle size
r = quantity measure

Calculation of the density distribution $q_r(x)$:

$$q_r(x) = \frac{\text{quantity } \Delta q \text{ (dq) of the measured particles in the intervall } \Delta x \text{ (dx)}}{\text{total quantity of the measured particles } q_{ges} \cdot \text{intervall width } \Delta x \text{ (dx)}}$$

The following is valid formally mathematically:

$$q_r(x) = \frac{dQ_r(x)}{dx} \qquad Q_r(x') = \int_{x_{min}}^{x'} q_r(x)dx$$

Quantity type: $r = 0$: number
$r = 1$: length
$r = 2$: face
$r = 3$: volume

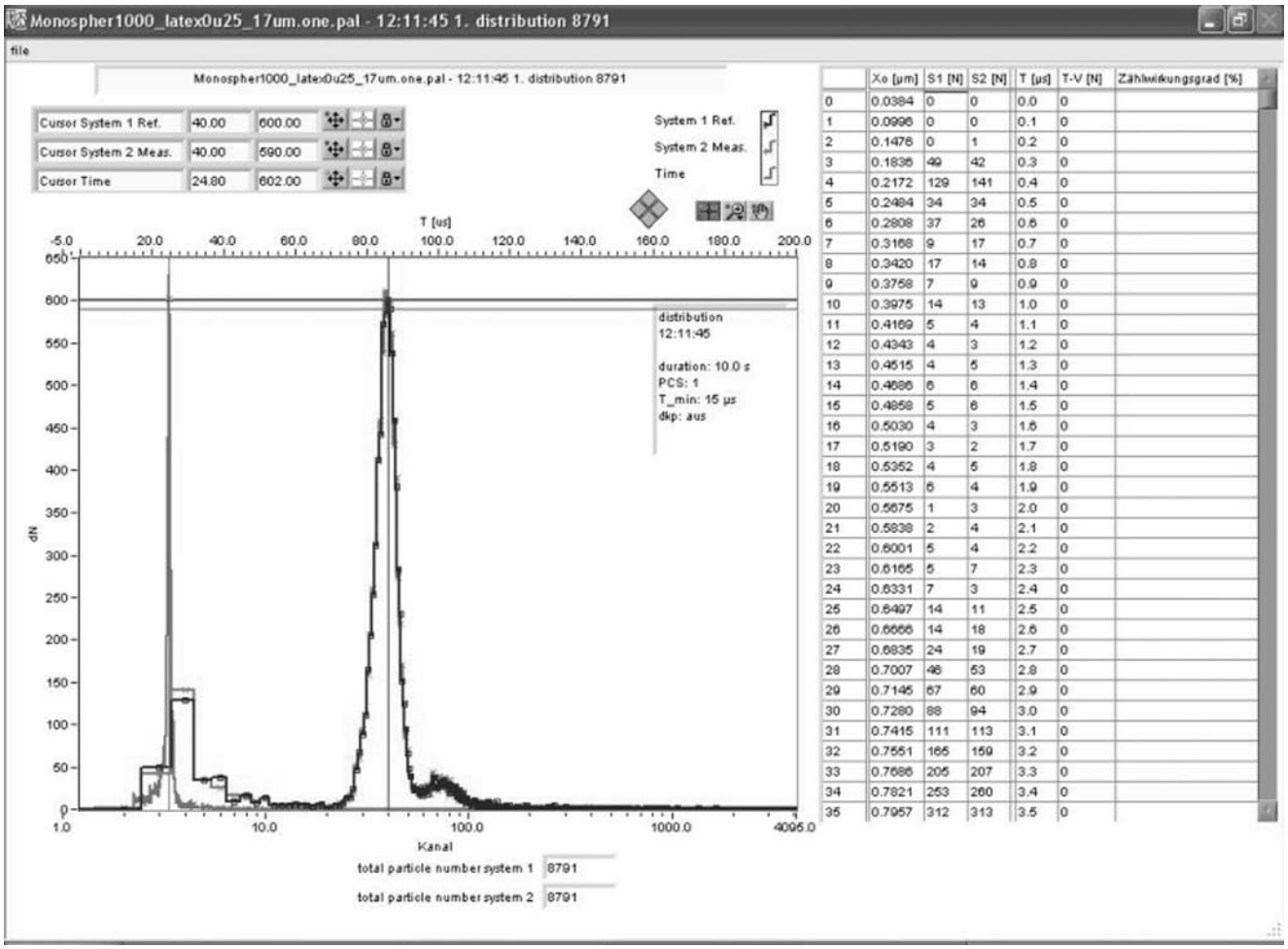

Figure 16:
Raw data analysis in the welas® software

5.2 Calibration

It was already mentioned how important is the calibration of a particle measuring instrument for a clear comparison of measurement results. Possibilities for the generation of test aerosols are represented in (Hochrainer S. 1998, Helsper C. et al. 1989, Peters C. et al. 1991).

Optical aerosol spectrometers are calibrated with latex (m = 1.59). One measures with such methods the scattered light equivalent diameters related to latex.

In the following, the importance of the correct calibration procedure will be shown considering as example the particle size characterisation of the aerosol spectrometer welas®.

As described in chapter 5, at the welas®-system – due to the evaluation of the flying time of the particles through the T-shaped measuring volume – all scattered light impulses are rejected automatically, whose evaluation with conventional measuring instruments would cause a border zone error. Due to the same technology, measurements in coincidence are indicated optically and/or acoustically.

For these reasons, the evaluation software of the welas® offers an analysis of the raw data concerning the particle size and particle velocity distribution as represented in figure 16.

The condition of the exact concentration determination is the accurate size of the measuring volume which is determined with a tolerance of ± 1.5 %.

Figure 16 shows the raw data saved during a measurement. One measured quasi monodisperse SiO_2-particles with a scattered light equivalent diameter of 0.87 µm related to Latex.

On the Y-axis the number of particles is represented. On the lower X-axis the raw data channels (raw size classes) are shown, on the upper X-axis the flying time in µs. In table of figure 16 one can read the corresponding values.

Since the welas®-system is supplied, depending upon application, with differently large measuring volumes and differently highly adjustable volume flows, the respective minimum permitted flying time T_{min} must be set by computer.

From the indication of the mean flying time of the particles and the length of the measuring volume one can clearly determine the mean particle velocity in the measuring volume for monosphere (figure 17).

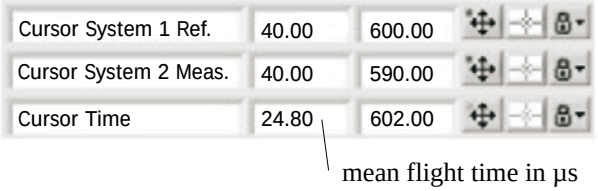

Figure 17:
Raw data analysis – detail

$$\bar{v}_{particle} = \frac{L_{measuringvolume}}{\bar{t}_{particle}}$$

$$\dot{V}_{measuringvolume} = A_{measuringvolume} \bullet \bar{v}_{particle}$$

$\bar{t}_{particle}$: *mean flying time of particles through the measuring volume*
$\bar{v}_{particle}$: *mean velocity of particles through the measuring volume*
$\dot{V}_{measuring\ volume}$: *volume flow in the measuring volume*
$A_{measuring\ volume}$: *passage surface of measuring volume*
$L_{measuring\ volume}$: *length of measuring volume*

Only the accurate determination of the particle velocity in connection with an accurate indication of the passage surface in the measuring volume leads to an accurate calculation of the measured particle concentration.

$$c_n = \frac{N_{analysed\ particles} \bullet t_{measuring\ time}}{\dot{V}_{measuring\ volume}}$$

c_n = *number concentration*
$N_{analysed\ particles}$ = *number of measured particles*
$t_{measuring\ time}$ = *measuring duration*

The raw data distribution is used also for the exact adjustment of the sensor with monodisperse particles. This calibration concerning the particle size can be accomplished by the customer himself.

5.2.1 Characterisation of the device parameters – border zone error, resolution and classification accuracy

The characterisation of the welas® 1200, which is equipped with a cuvette, was accomplished in the Leibniz-Institut für Troposphärenforschung e. V. (IFT) in Leipzig, Germany. In the figures 18 and 19 the measuring data of three different latex aerosols are represented. The latex suspensions (0.2 µm; 0.34 µm; 0.5 µm) were generated in each case in a separate Palas® latex generator and were supplied directly to the welas® system.

It can be clearly taken from the run of the cumulative distribution in figure 19 that besides latex there are also finer particles in the aerosol. As it is well known, one does not receive an exclusive mono-disperse aerosol when nebulising latex suspensions, since the stabilisers of the suspension are also nebulised and measured. This fact supplies an explanation for the fines recognisable in figure 19. However, it can not be excluded that the fines represented in the curve could also be due to a remainder border zone error of the measuring system.

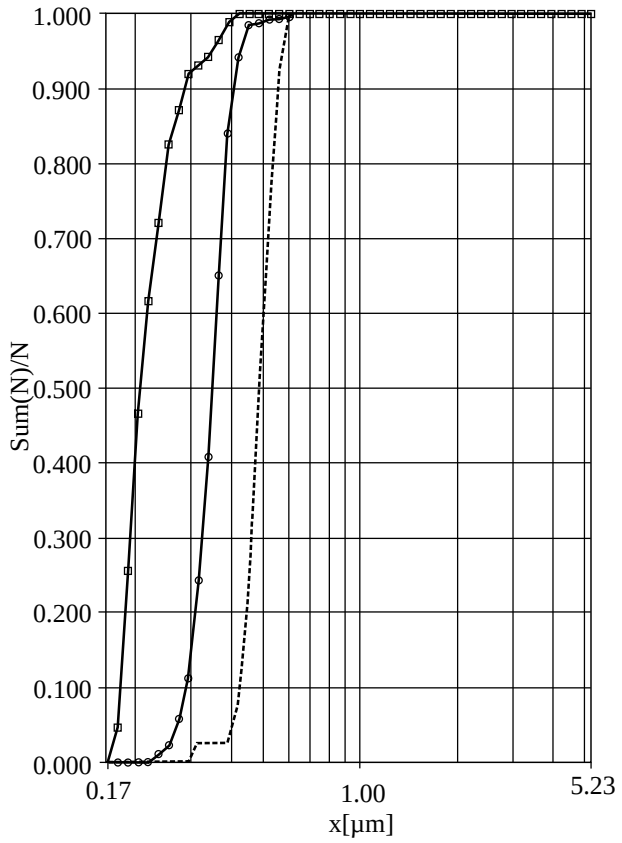

Figure 18:
Latex aerosols dp = 0.2; 0.34; 0.5 µm with fines separated by a DMA

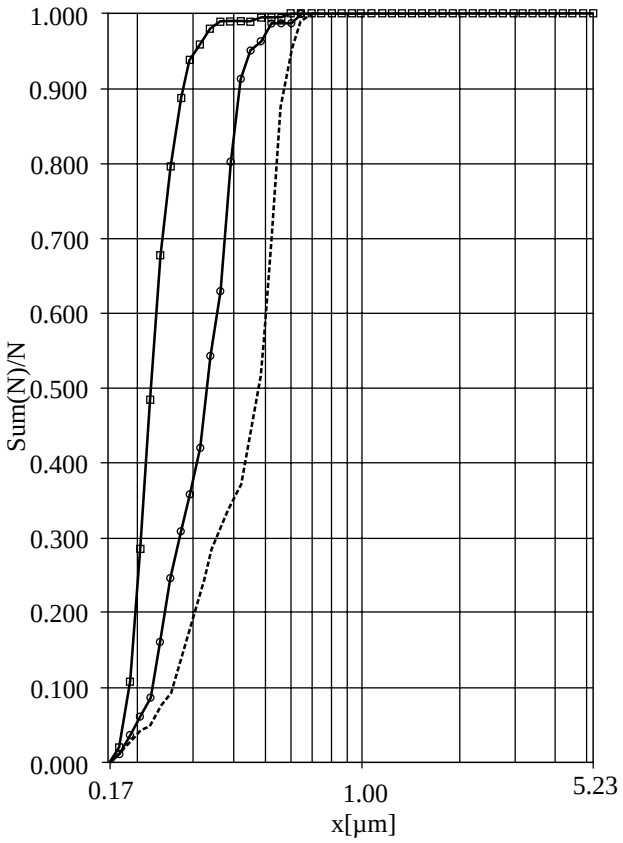

Figure 19:
Latex aerosols dp = 0.2; 0.34; 0.5 µm with fines nebulised by the stabilisers of the suspension

For the clear characterisation of the aerosol spectrometer, the three different latex aerosols were led through a company-owned Differential Mobility Analyser (DMA) of the IFT. The DMA was switched between the latex generator and the welas®-system. In figure 18 it can be clearly recognised that the fines of the aerosol were separated by the DMA and that they are therefore not measured and indicated by the welas®-system. Thus, it can be concluded that the T-aperture-technology supplies practically border-zone-error free measurement results. Therefore, the curve in figure 19 was measured correctly with the welas®, since the latex aerosol was soiled with fines. The curves show clearly the high resolution and the good classification accuracy of the welas®-system. One recognises the resolution by the rate of rise of the cumulative distribution. The classifying accuracy is read off from the d_{50}-value of the cumulative distribution.

6 Summary and future prospects

Counting optical aerosol spectrometers are suitable for the meaningful particle size and particle number determination; they must however fulfil certain technical conditions. Among them there are the high resolution and the good classification accuracy, i.e. a measuring method which has a clear calibration curve and measures without border zone errors. Coincidence-free measurements are ensured by coincidence detection.

The insight into the theoretical basics of the counting optical particle measurement explains the significance of the device parameters and the importance of a clear calibration curve. The explanation of the operational principle of optical measuring methods demonstrates the advantages of a measurement with white light and 90° scattered light detection compared to laser light or forward scattering.

The white light source and the 90° scattered light detection provide a clear calibration curve. A specific feature is the patented T-aperture-technology: The T-shaped measuring volume projected and limited with white light allows for the first time a border-zone-error free measurement.

In addition, the measuring system possesses coincidence detection. The heatable measuring chamber and the optical fibre technology offer special advantages, too. Due to the modular set-up the sensors with the optimal measuring volume size can be selected depending upon application. Beyond that, the welas®-system offers an innovative specific feature which pays off particularly with special applications, for example measurements in strongly varying or very high raw gas concentrations or measurements in badly accessible environments: For the first time one can measure with two sensors at one control unit quasi simultaneously at different places in different concentrations.

The evaluation of the scattered light signals generated by the particles is done by software tested in practical applications. This software does not only allow different representation possibilities of the particle size distribution, but also records the environment conditions such as air pressure, relative humidity, temperature and velocity, which were measured with other sensors. The data can be recalled also over the internet.

An important advantage of the welas®-system is - in particular also with view to the future - the possibility to combine the measurements with other measuring methods (Mölter L. et al. 2004).

7 References

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Particle size and shape distribution of stable dust analysed with laser diffraction and imaging technique

M. Romann[1] and T. Hinz[2]

Abstract

Effects of particles on individuals and the dispersion of airborne particles are strongly dependent on their size, density, surface and shape. Risk assessment requires knowledge about mass or number concentration or fluxes concerned. For this purpose different kinds of measuring techniques are used, which react in particular way to the parameters mentioned above. Depending on the task an appropriate technique will be selected. It's well known that dusts from livestock facilities vary widely depending on the source. Sophisticated structures and large ranges of size distribution must be detected. For sizing laser diffraction technique was used to characterize samples of dust which were collected in stables of different species of farm animals. These results are now supplemented with Sympatecs imaging system for shape analysis.

Keywords: livestock, PM, size, shape, laser diffraction, high speed imaging

Introduction

Effects of particles on individuals and the dispersion of airborne particles are strongly dependent on their size, density, surface and shape (Schmitt-Pauksztat G. 2006). Depending on these parameters particles penetrate more or less deep into the breathing of individuals. Aerodynamic diameter is used to cover complex influences including shape and structures. This becomes complicated for particle dispersion caused by emissions in animal production. Therefore the well established laser diffraction technique for analyzing particle size distribution is not sufficient. In the following more detailed parameters for the characterization of the dust particles are described.

Materials and Methods

Particle analysis was carried out in the labs of Sympatec with samples which were taken in research facilities of the FAL and in a commercial piggery near Braunschweig. The different dust types are taken in stables of cattle, piglets, pigs, horses, sheeps and turkey.

The collection location was a central point inside of each stable with the exception of turkey where the sample was taken from the emission flow. This type of stable dust was chosen for the first investigations.

According to the conditions of measurements at working places (VDI 1980) probes were sucked with a velocity of 1.25 m/s in a height of approximately 1.5 m above ground. Figure 1 shows the setup of the sampler. Samples in the exhaust of the force ventilated turkey kept the conditions of isokinetic probe (DIN EN 12384-1, ISO 7708).

Independent of the air intake the equipments consist of a high volume sampler with an axis-type cyclone to separate the coarse fraction and a glass fibre filter to collect the penetrating fine dust particles, figure 2.

[1] Sympatec GmbH, Clausthal-Zellerfeld, Germany

[2] Federal Agricultural Research Centre, Institute for Technology and Biosystems Engineering, Braunschweig, Germany

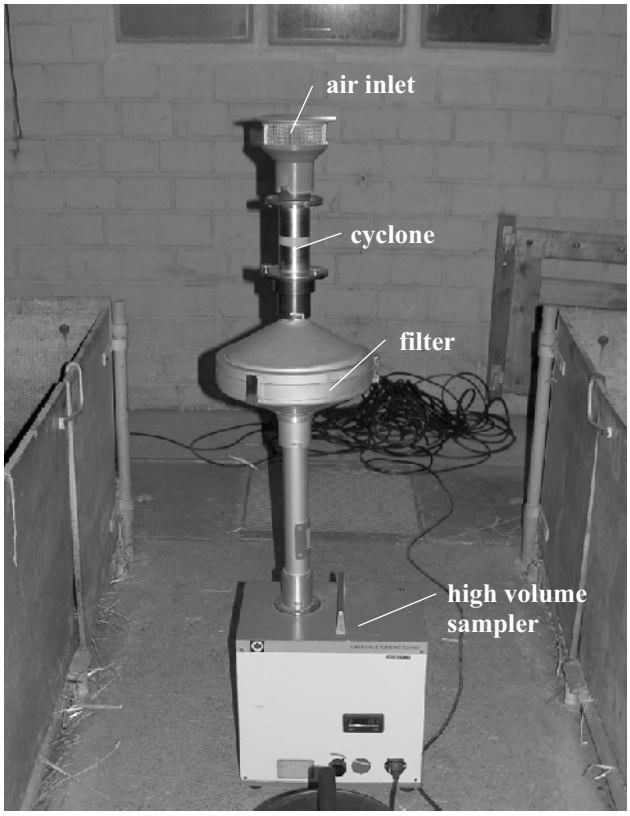

Figure 1:
Sampling dust inside a stable

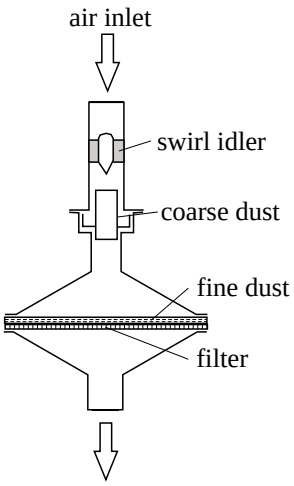

Figure 2:
Scheme of the equipment

The separation efficiency of the cyclone was fitted to the previously used Convention of Johannisburg, with a cut off diameter of 5 µm. Particles larger than 7.07 µm should be separated totally. Figure 3 shows the theoretical curve and the actual calibration while sampling with a flow rate of 50 m³/h.

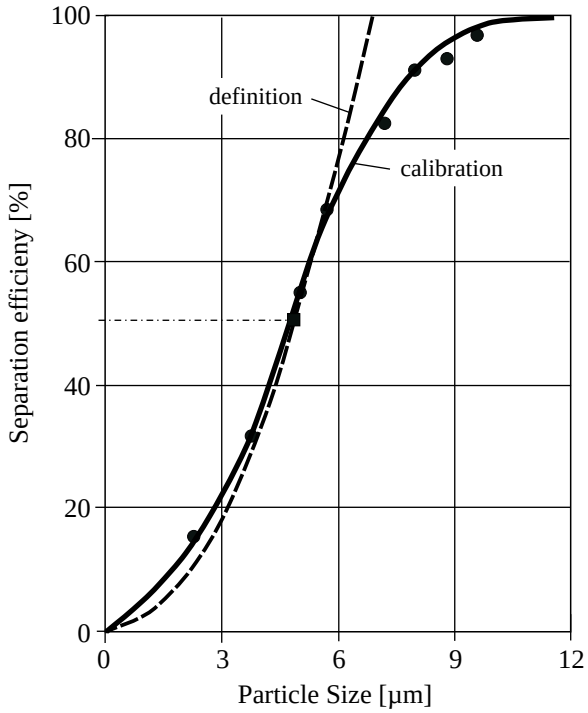

Figure 3:
Separation efficiency of the used cyclone

High volume sampling ensures proper amounts of dust in the cyclone beaker and on the filter in an acceptable time of running. Mass detection followed by weighing. After weighing the samples of the coarse fraction were transferred to the lab for further analysis.

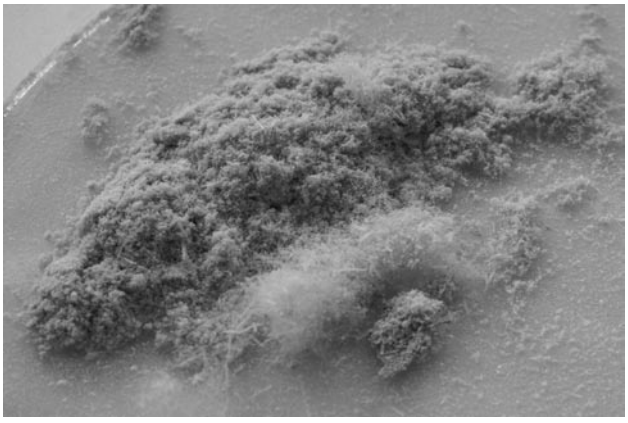

Figure 4:
Sample of stable dust

This sample can be divided into two different types of particles: a dusty appearing collection of “normal” particles and balls of fibrously particles that can be taken out by tweezers. A dry sample splitting is impossible.

The best way to handle the sample of stable dust carefully is to prepare a suspension of the entirely sample in dry isopropylic alcohol. Stirrer speed should be slow and it should change the rotation direction from time to time in order to avoid the collecting of fibres or the creation of fiber balls again.

The suspension can now be split and diluted for the measurements first with a HELOS laser diffraction together with the dispersing unit SUCELL and then with a QICPIC imaging system combined with the dispersing system LIXELL.

Results and discussion

Table 1 gives the concentration of the coarse and fine fractions split by the cyclone separator and its ratio coarse to fine. The suspension made up for the particle size distribution analysis and for the shape analysis contains only the coarse fraction.

Table 1:
Concentration of coarse and fine dust fraction, percentage coarse

Species	Fraction concentration coarse [mg/m³]	fine [mg/m³]	Percentage coarse	Position
Cattle I	0.116	0.00567	96	inside
Cattle II	0.180	0.029	87	inside
Piglet	1.144	0.159	88	inside
Pigs	0.860	0.057	93	inside
Horse	0.150	0.0268	85	inside
Sheep	0.072	0.017	89	inside
Turkey	2.560	0.296	90	exhaust

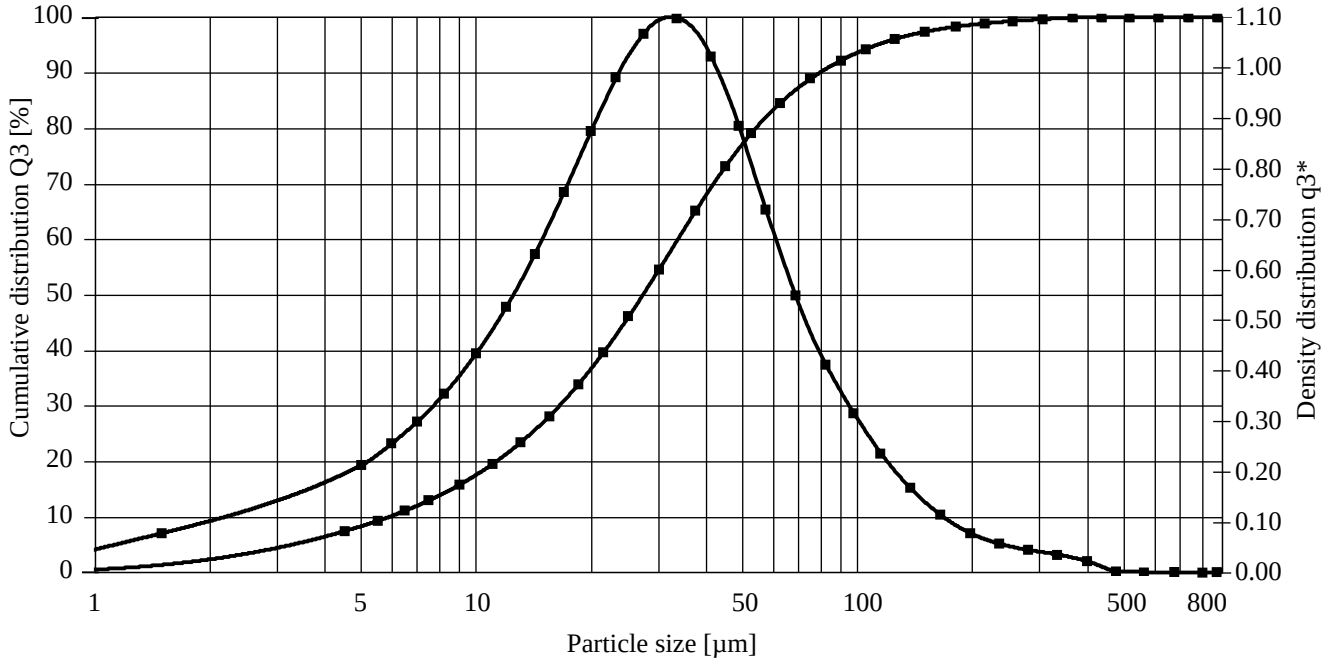

Figure 5:
HELOS - Cumulative and density distribution of the turkey stable dust.

The result of the laser diffraction analysis shows a distribution of laser diffraction comparable spheres. The measurement itself is done in less then one minute and is based of some million particles. Therefore the reproducibility and reliability of the result are very high. The standard deviation of three different measurements is below 0.5 %.

But even the best result does not show any information about particle shape.

More detailed information about the particles is given by the imaging system QICPIC. Its singularly combination of a high speed camera up to 500 images per second, shortest exposure times below 1 ns with special telecentric optics makes it possible to evaluate a high number of sharp particle images (Witt W. et al. 2006 and 2007).

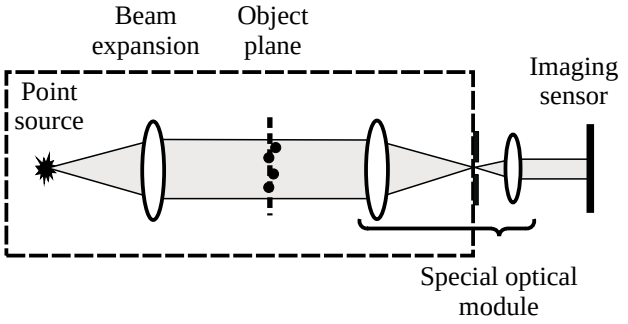

Figure 6:
Optical setup of QICPIC imaging system.

Before and during the measurement the sensor-control window of the WINDOX-software shows already particle images. This allows to check the measuring range and the concentration of particles in the suspension.
Considering the flow velocity in the cuvette and the volume of the suspension a frame rate of 50 images per second during a measuring time of 120 seconds has been suitable. In this case the measurement contains the images of 350,000 particles.

The software WINDOX calculates parameters like EQPC (diameter of the circle of equal projection area), the different Feret-diameters (distance between to parallel tangents), the minimum bounding area with its maximum and minimum value, length and diameter of fibres and the volume based fibre diameter. This list has to be supplemented with the shape parameters like sphericity, aspect-ratio, convexity, straightness and elongation of fibres and with the possibility to combine these parameters to USER-defined parameters.

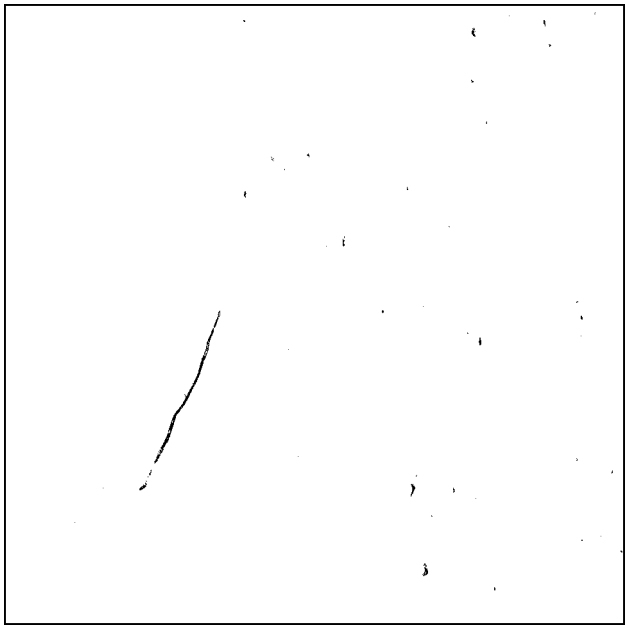

Figure 7:
QICPIC - Particles flowing through the cuvette during the measurement

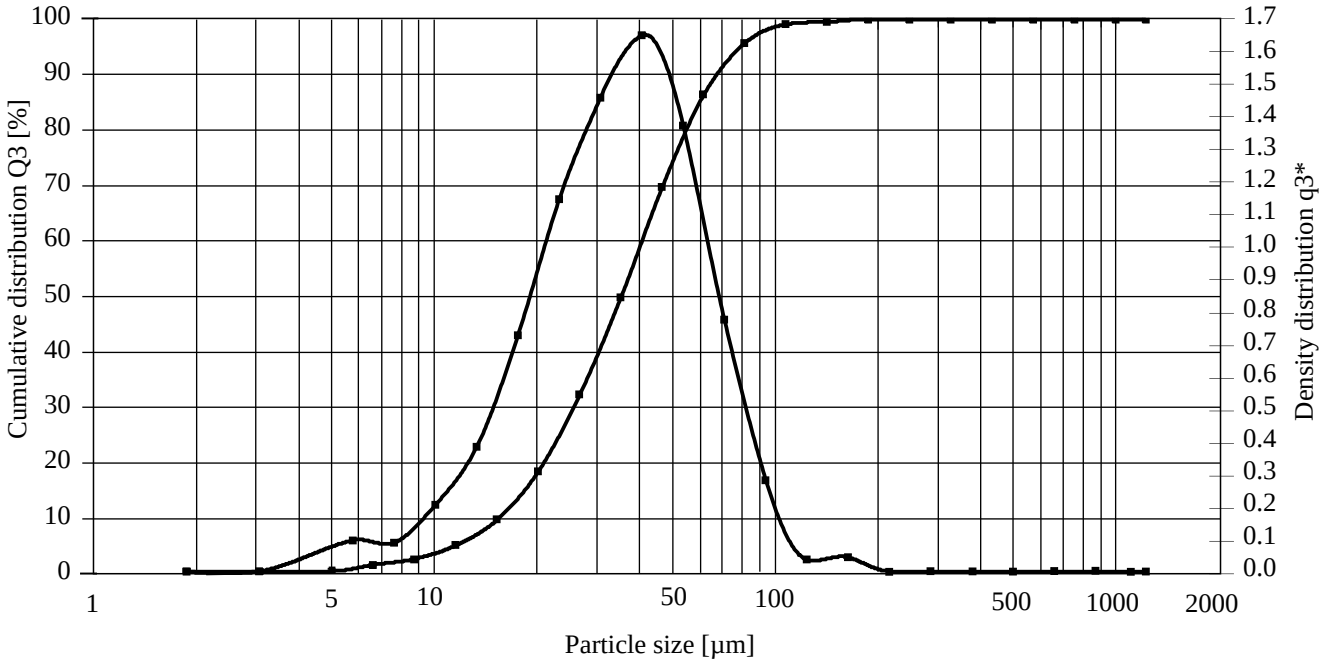

Figure 8:
QICPIC - Volume distribution of EQPC as first result. This diagram is slightly comparable to the volume distribution diagram of the laser diffraction result. (Figure 4)

With all these parameters and their combinations a widespread variety of different results can be created to characterize the very special properties of the sample:

- Particle size distribution of different particle diameters
- Distributions of user defined particle parameters
- Shape diagrams of different shape parameters vs. particle size
- Distribution diagrams of different shape parameters
- Shape diagrams of USER-defined particle parameters

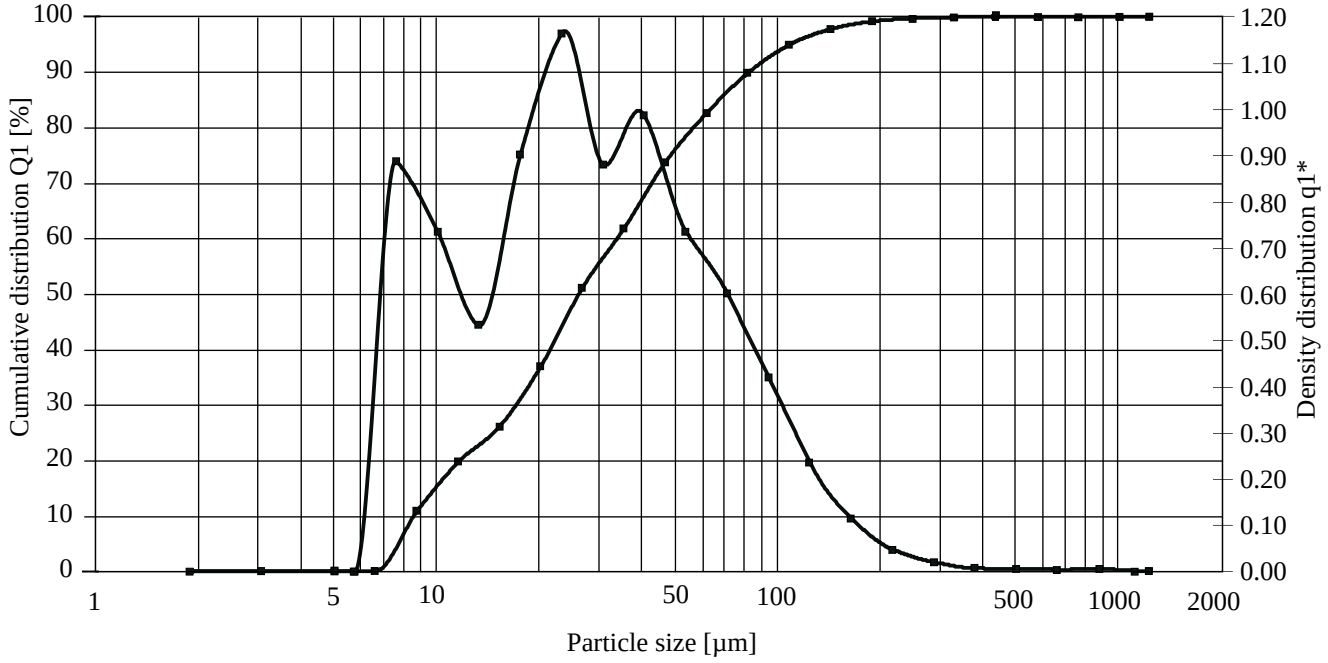

Figure 9:
QICPIC - Length distribution of Feret-Max as result for fibrously materials

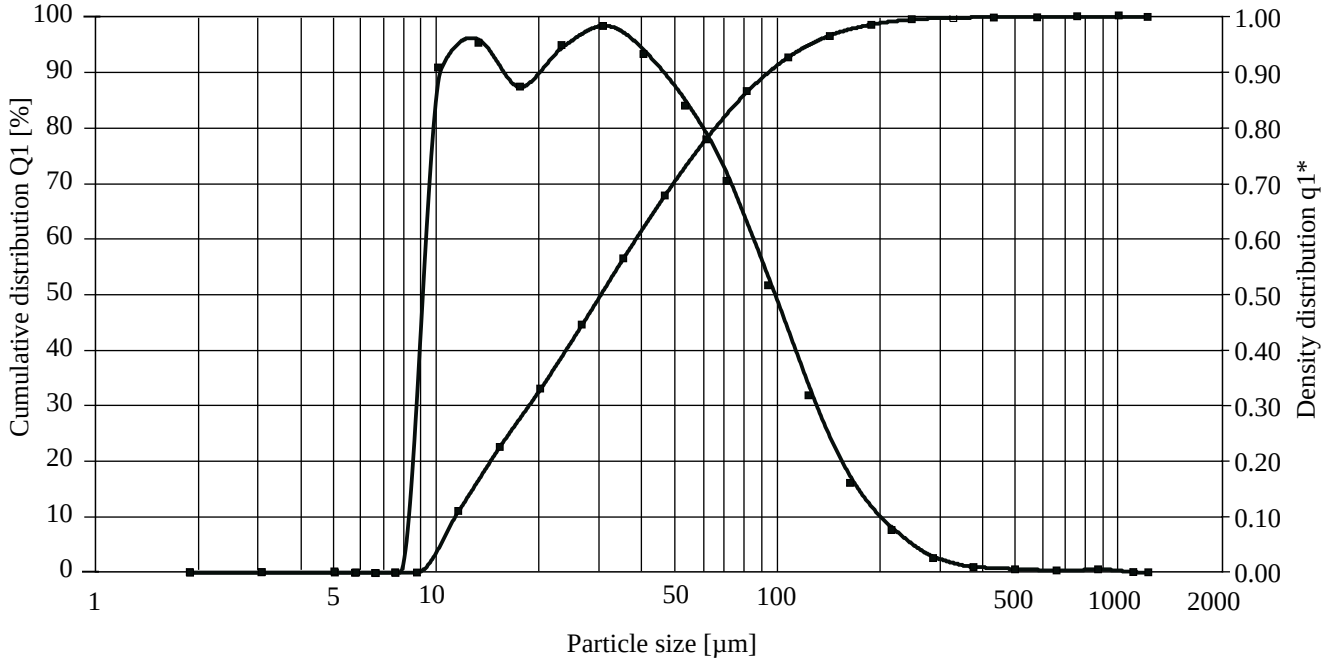

Figure 10:
QICPIC - Length distribution of LEFI (Length of Fibre)

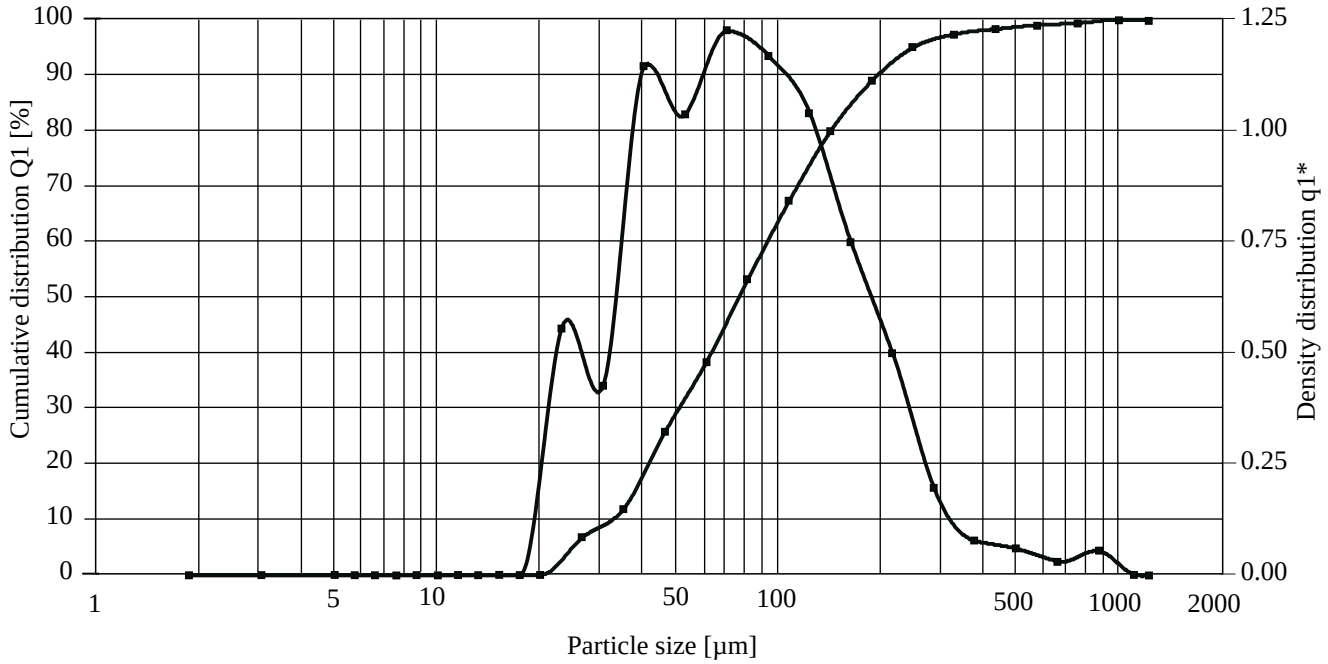

Figure 11:
QICPIC - Length distribution of LEFI > 20 µm and Aspect ratio < 0.25. This user filter suppresses the part of small, spherical particles

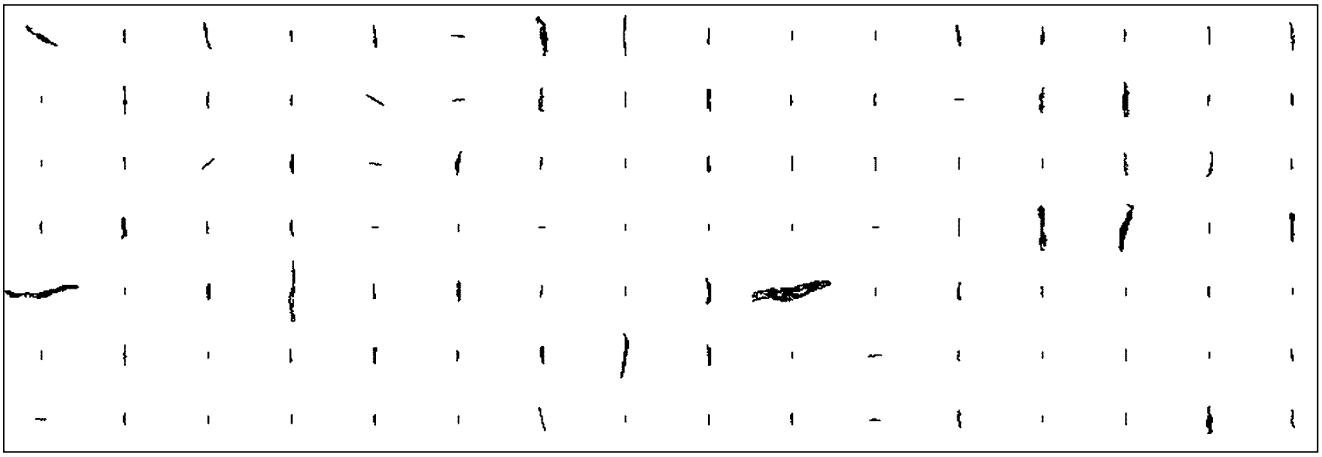

Figure 12:
QICPIC - Particle Gallery of the distribution shown in Figure 10

The last hint, that the sample of stable dust contains mostly needle-shaped particles is found in a shape-diagram:

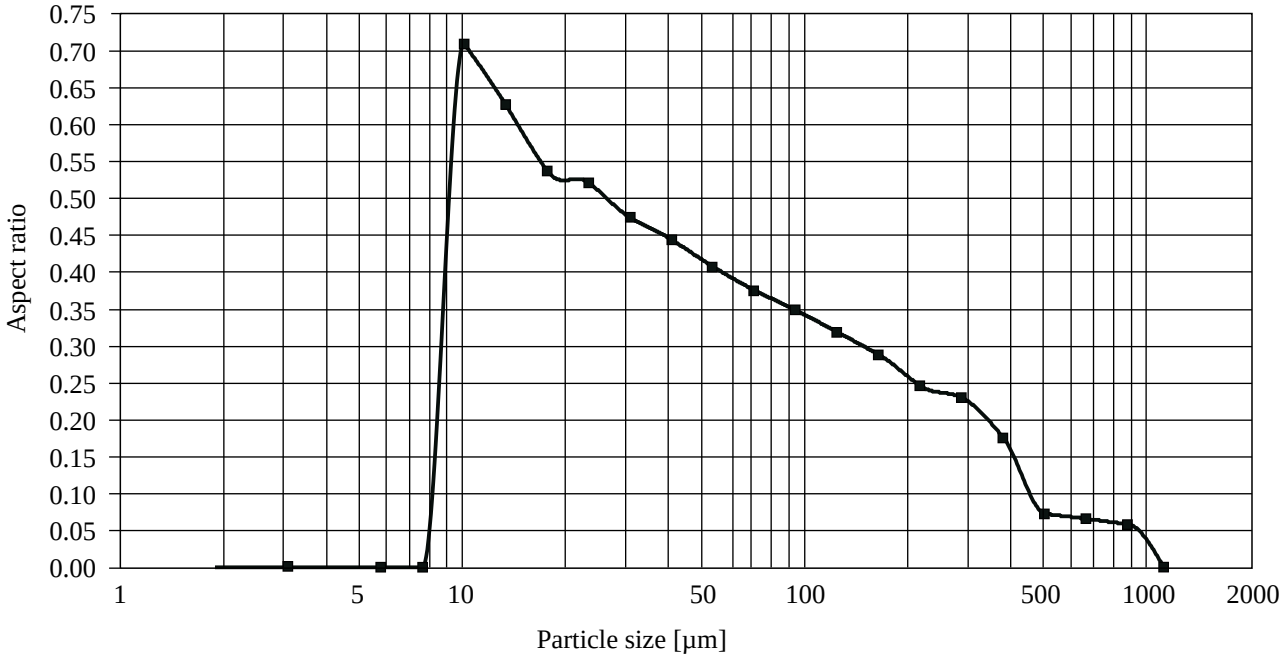

Figure 13:
QICPIC - Aspect ratio vs. LEFI. Typical for fibrously samples is the falling curve to bigger particles

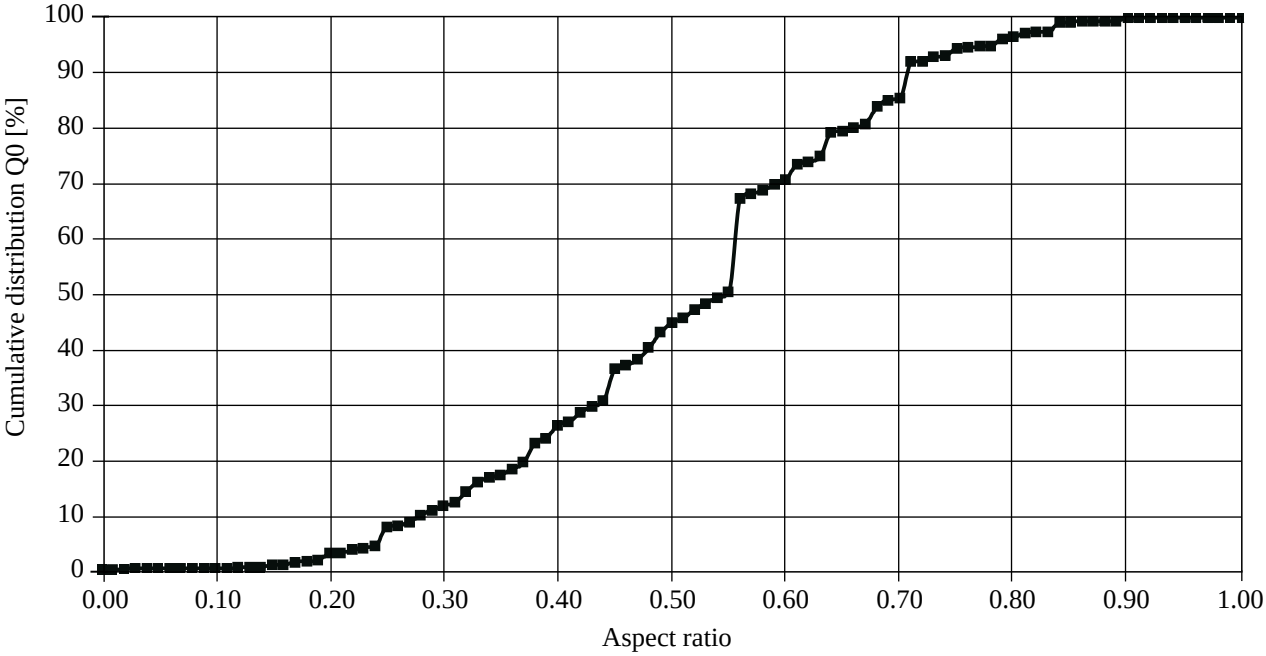

Figure 14:
QICPIC - Cumulative number distribution of aspect ratio. A flat curve shows the predominantly influence of fibres

Conclusion

First investigations in stable dust dispersed as suspension in dry isopropylic alcohol, show very positive results with Sympatec laser diffraction system HELOS with SUCELL that allow a suitable handling of this special material and gives reproducible results of particle size distribution. This realization has been transferred to the imaging system QICPIC with LIXELL. Its variously possibilities in the evaluation of stable dust lead from simple particle distributions in order to have an overview down to the discovering of single particles with very special user specified properties.

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For detailed informations about the laser diffraction system HELOS and the imaging system QICPIC please refer to www.sympatec.com (english side) or www.sympatec.de (german).

Emissions of particulate matter from diesel engines - Determination of the particle number concentration in diesel exhaust gas and - Emissions of heavy-duty diesel engines with focus on particulate matter

Yvonne Ruschel[1], Olaf Schröder[1], Jürgen Krahl[1,2], and Axel Munack[1]

Abstract

Particulate matter (PM) has become perceived as one of the major harmful emissions from diesel engines. PM emissions are subject to diesel engine emission legislations worldwide. Epidemiological and toxicological studies have indicated that adverse health effects increase with decreasing particle size and increasing particle number. Therefore, the determination of the particle number distribution is of same importance as the assessment of the total mass of PM. In the first part of this paper, measurement techniques of particulate matter and two instruments for determination of particle number concentration and particle size distribution in diesel exhaust gas are introduced. The second part gives an overview of particulate emissions of a heavy-duty diesel engine running on four different fuels.

Keywords: Biodiesel, diesel fuel, particulate matter, ultrafine particles, health effects, ELPI, SMPS

Introduction

Particulate Matter (PM) has become perceived as one of the major harmful emissions from diesel engines. PM emissions are subject to diesel engine emission legislations worldwide. According to the U.S. Environmental Protection Agency (EPA), particles are defined as all solid or liquid substances in diluted exhaust gas that can be collected on a filter at a temperature of under 51.7 °C (which is 125 °F) (Code of Federal Regulations). The temperature reduction of the exhaust gas is achieved by diluting the exhaust gas with air.

PM consists of a variety of organic and inorganic substances. The main constituents of the organic substances are unburned or partially burned fuel and lubrication oil compounds. The inorganic substances include soot (carbon), sulphates, water and metallic compounds. Metal and rust particles from the engine or the exhaust gas system as well as derivates of organo-metallic fuel and lubricant additives are included in the metallic compounds. The percentage of these substances in the total particle mass depends on a multitude of parameters. In addition to constructive parameters such as design of the combustion chamber and the injection system, the mode of operation, or rather the overall load configuration, the fuel and lubricant quality as well as the wear of the engine are also included here (Wachter F. and Cartellieri W. P. 1987).

Recently emissions of fine and ultrafine particles from diesel engines induced a broad discussion in Germany. Since January 2005 the limit for the emission of fine particles has been tightened in the context of a council directive of the European Union (99/30/EG).

According to this directive the limit of 50 µg/m³ particles in ambient air should not be exceeded. The compliance of the limit cannot be guaranteed yet all over in Germany.

In 2006, an exceedance of the limit on more than 35 days per year occurred at 100 of the 450 measuring stations.

In detail, particles from diesel engines show a bimodal size distribution consisting of a nuclei mode and an accumulation mode as can be seen in figure 1.

[1] Federal Agricultural Research Centre, Institute for Technology and Biosystems Engineering, Braunschweig, Germany

[2] University of Applied Sciences Coburg, Coburg, Germany

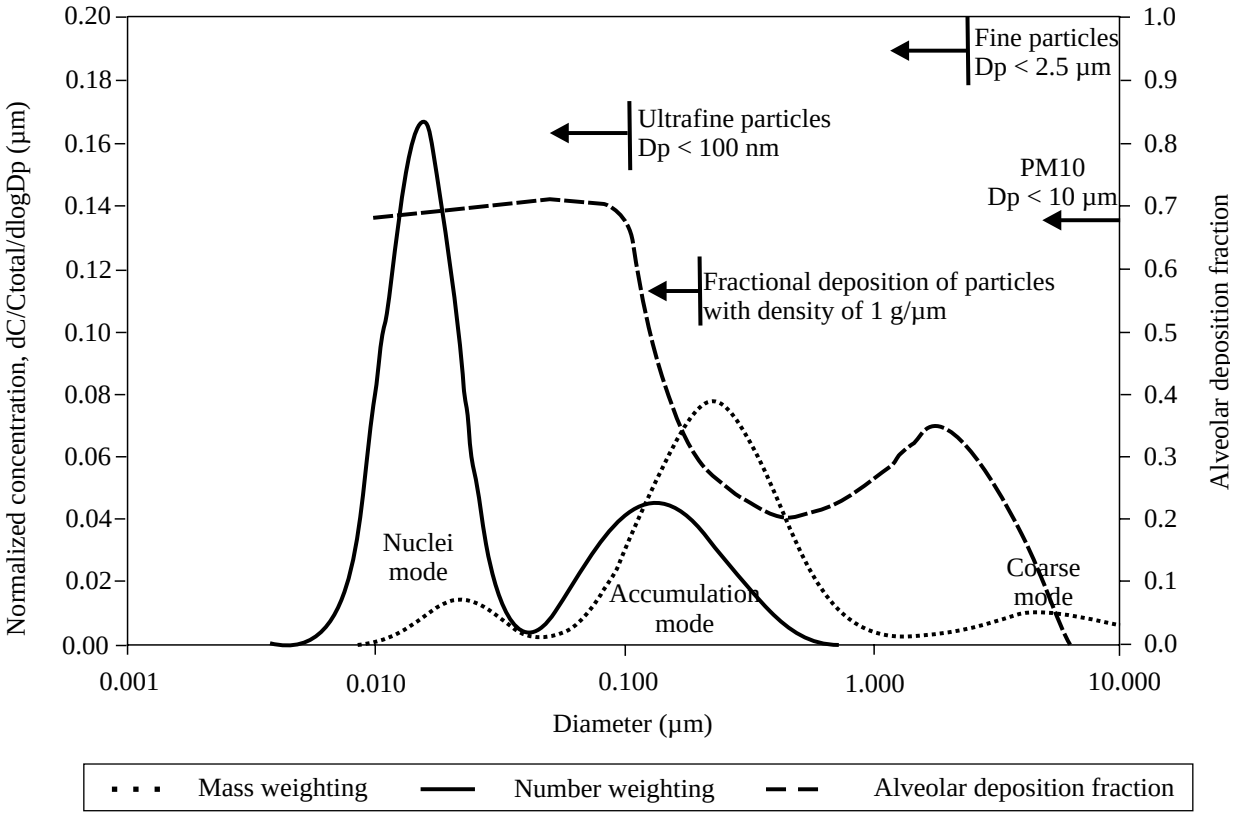

Figure 1:
Typical diesel engine exhaust particle distribution by mass, number and alveolar deposition (Kittelson D. B. 2000)

Nuclei mode particles, which can depose deep in the lungs, dominate the majority of particles but have only little effect on the total mass. Epidemiological and toxicological studies indicated that adverse health effects from exposure to PM may increase with decreasing particle size even if the particles consist of toxicologically inert materials (Seaton A. et al. 1995, Donaldson K. et al. 2000). Ultrafine particles can penetrate the alveolus region, resting there for months. Here the particles can cause allergic or inflammatory reactions leading to bronchitis and asthma (Mayer A. 2001). Therefore the determination of the particle number distribution is just of the same importance as the PM mass that is subject to regulation. Consequentially, the particle number is currently under discussion as future regulated value for Euro 6 legislation.

Beneath results on emissions this paper presents briefly two instruments for the determination of particle number concentration in diesel exhaust gas as well as the sampling procedure for PM. A scanning mobility particle sizer (SMPS) detects particles based on their electrical mobility. SMPS enables the determination of particles in the size range between 10 to 300 nm. A low pressure impactor (ELPI) detects particles between 30 nm and 10 µm. Furthermore, the overall particulate mass (PM) can be determined gravimetrically.

The presented particulate emissions data of a heavy-duty diesel engine running on four different fuels were obtained be applying all three measurement methods.

Material and methods

Test Engine

Studies were carried out at the emission test stand of the Institute for Technology and Biosystems Engineering at the Federal Agricultural Research Centre (FAL) in Braunschweig, Germany. A Mercedes-Benz engine OM 906 LA (Euro 3) with turbocharger and intercooler was used. The engine was equipped with a pump-line injector system. It was operated without exhaust gas recirculation. Table 1 presents some of the engine characteristics.

Table 1:
Technical data of the test engine Mercedes-Benz OM 906 LA

Piston stroke	130 mm
Bore of cylinder	102 mm
Number of cylinders	6
Stroke volume	6370 cm^3
Rated speed	2300 min^{-1}
Rated power	205 kW
Maximum torque	1100 Nm at 1300 min^{-1}
Compression ratio	17.4

Exact engine load during test cycles is guaranteed by crankshaft coupling to a Froude Consine eddy-current brake. Engine test runs were in accordance with the 13-mode European Stationary Cycle (ESC), for which the preset torque and revolution rates (related to maximum load at the actual speed or related to rated speed, respectively) as well as the time courses of engine torque and speed are displayed in subsequent figures 2 and 3.

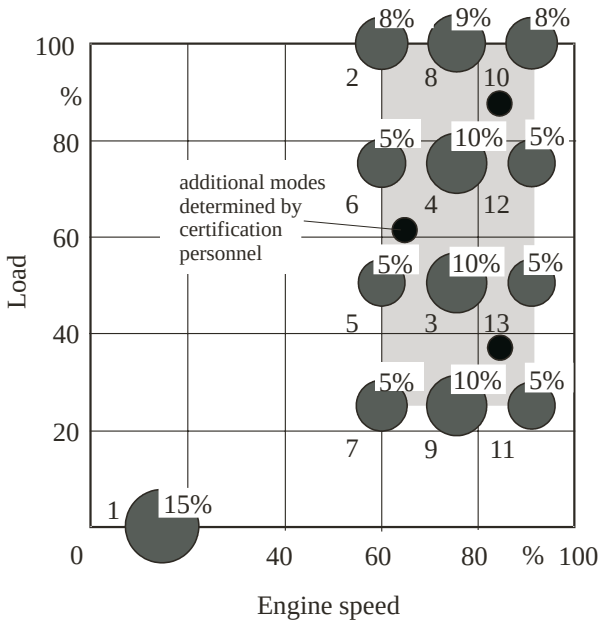

Figure 2:
Modes of the ESC test cycle

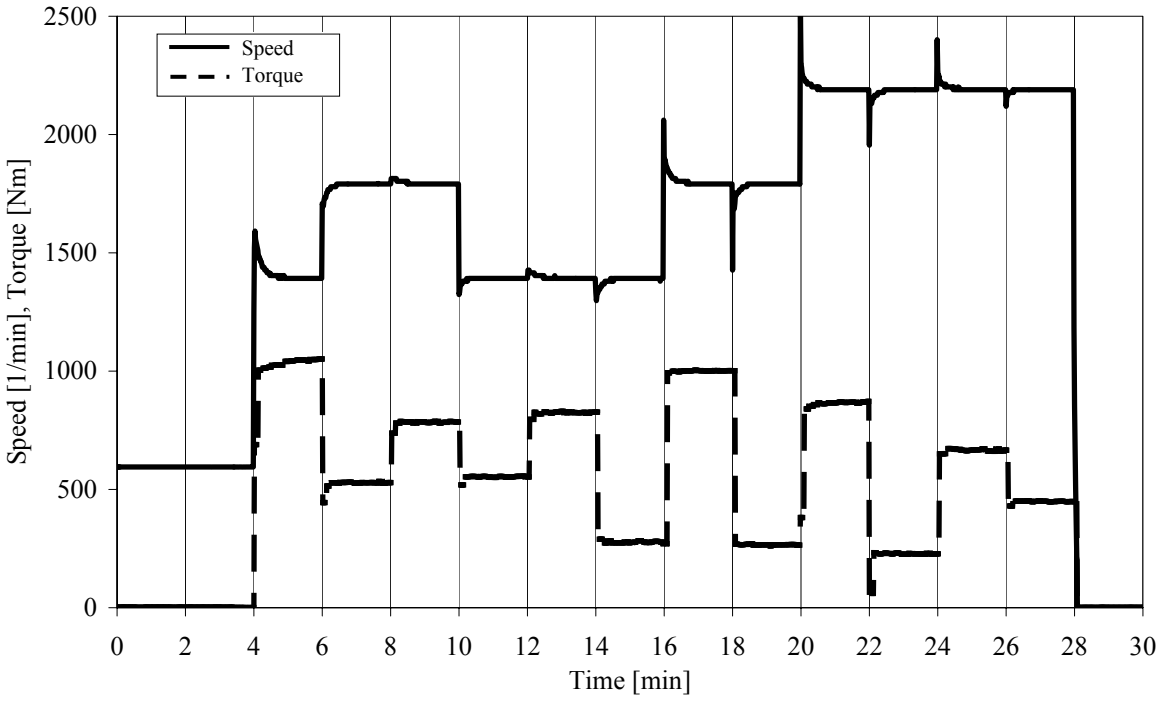

Figure 3:
Exemplary courses of engine speed and torque for an ESC test cycle

In previous investigations higher emissions of particles in the nuclei mode were detected for RME compared to other fuels (Munack A. and Krahl J. 2005; Herbst L. et al. 2006; Ruschel Y. et al. 2006). Therefore, RME was compared to two common diesel fuels and one artificial blend. The following four fuels were used for engine operation:

DF:	Reference diesel fuel
RME:	Rapeseed oil methyl ester
B5Ult:	5 % RME + 95 % Aral Ultimate Diesel®
V-Power:	Shell V-Power Diesel®

RME was provided by Carl Büttner Mineralöl-GmbH in Leer; DF from Haltermann Products BSL Olefinverbund GmbH in Hamburg. Aral Ultimate Diesel® and Shell V-Power Diesel® were obtained from public filling stations in Braunschweig.

Particulate measurement

The emissions of particles were determined by three different ways: The total mass of particulate emissions was measured gravimetrically according to the EU regulation (EU, 2005). Particle number and particle size distributions were measured with ELPI and SMPS.

Particulate Matter

PM measurements were accomplished after partial flow dilution of raw exhaust gas in a dilution tunnel (figure 4), which cools down the exhaust gas to under 51.7 °C. A dilution factor of about 10 is applied for determination. Dilution factors are calculated from separate recordings of CO_2 contents in exhaust gas, fresh air, and diluted exhaust gas.

The particles are collected on a two stage filter. Particle mass was determined gravimetrically after deposition to teflon-coated glass fibre filters (T60A20, Pallflex, diameter 70 mm).

The total volume (V_{SAM}) that is led through the filter results from the requirement that the sampling time for each testing phase must be at least four seconds per 0.01 weighting factor. This must also take place as late as possible and may not be concluded earlier than five seconds before the end of the phase. For sampling a mass flow controller is set in such a manner that it is in accordance with the individual weighting factors of each mode of the ESC test. Each sampling takes 60 seconds and ends three seconds before the end of the mode. In this time the dilution level is constantly monitored and the exhaust gas sample volume can be corrected through a lengthening or shortening of sample time if the dilution factor changes. At the end the weighting factor that results from the following equation must be kept at ± 7 % to the weighting factor of the ESC-test.

$$\frac{V_{SAM,i} \cdot \sum_i \left(V''_{EDF,i} \cdot WF_i\right)}{V_{SAM} \cdot q_i \cdot V''_{EDF,i}} \overset{!}{=} WF_i$$

with

V_{SAM}:	Total volume of samples
$V_{SAM,i}$:	Volume of sample in mode i
$V''_{EDF,i}$:	Volume flow of exhaust gas in mode i
WF_i:	Weighting factor of mode i
q_i:	Exhaust gas dilution factor in mode i.

Weights of fresh and sampled filters were determined to an accuracy of ± 1 µg by means of a microbalance M5P from Sartorius, always preceded by at least 24 hours of

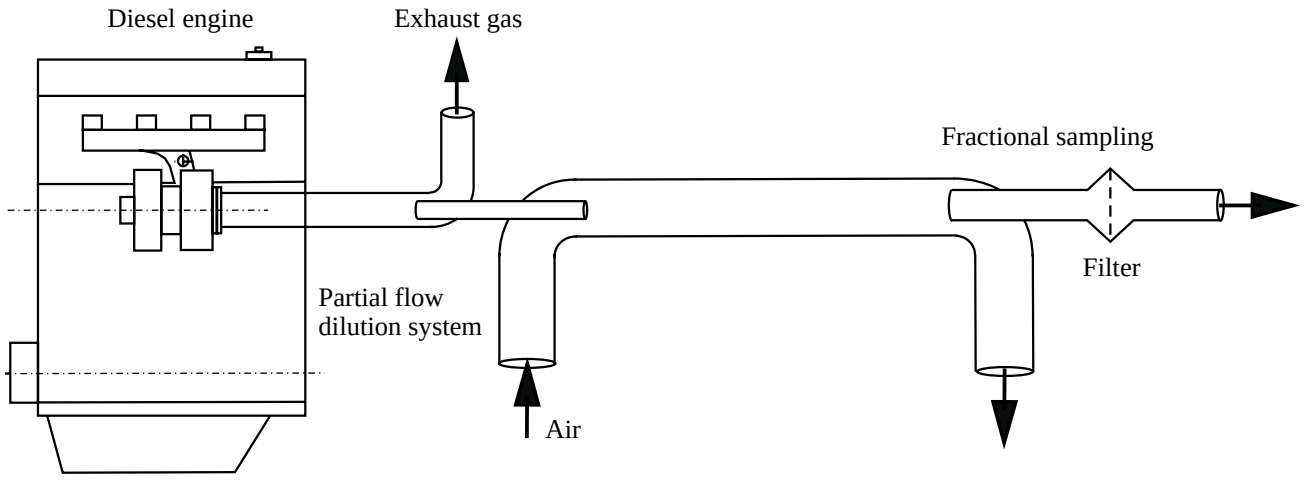

Figure 4:
Schematic diagram of the exhaust gas dilution tunnel

conditioning in a climate chamber held at 25 °C and 45 % relative humidity.

The particulate emission can then be calculated as

$$PT = \frac{M_{PF} \cdot \sum_i \left(V''_{EDF,i} \cdot WF_i \right)}{V_{SAM} \cdot \sum_i \left(P_i \cdot WF_i \right)}$$

with

PT:		Specific particulate emission
M_{PF}:		Total mass on the particle filter
V_{SAM}:		Total volume of samples
$V''_{EDF,i}$	:	Volume flow of exhaust gas in mode i
WF_i:		Weighting factor of mode i
P_i:		Power in mode i.

Particle size and number distribution

Determination of the particle size distribution is carried out by sampling with a multi-hole probe at the end of the exhaust gas dilution tunnel using a scanning mobility particle sizer (SMPS) system from TSI company. Secondary dilution is required to avoid an overloading of the measurement instrument.

The SMPS consists of a Kr-85 bipolar charger, an electrostatic classifier, both placed in the differential mobility analyzer (DMA), and a condensation particle counter (CPC). Before the sample aerosol flows through the DMA, particles larger 300 nm are removed by an impactor which is located in front of the DMA. The polydisperse aerosol is first led to a Boltzmann charge distribution by beta emission from the ^{85}Kr source (Liu B. Y. H. and Pui D. Y. H. 1974; Wiedensohler A. 1988). After passing the charger the aerosol is conducted into the electrostatic classifier. Figure 5 shows a general scheme of the DMA.

The electrostatic classifier consists of two concentric cylinders. The outer cylinder is grounded. Between the cylinders flows a sheath gas of filtered air. The charged sample aerosol is introduced along the inner wall of the outer electrode. The positively charged particles are accelerated toward the inner electrode by the electric field. As a result of the deflection the positively charged particles are dragged through the sheath air. Consequently, they quickly reach a terminal radial velocity that depends on the electrical mobility of the particles, that is independent of mass and density (Willeke K. and Baron P. A. 1993, Maricq M. M. et al. 2000, Hinds W. A. 1989). Depending on the voltage applied across the cylinders, only particles within a narrow range of electrical mobility, and therefore diameter, can pass through the exit aperture of the classifier (Knutson E. O. and Whitby K. T. 1975, Maricq M. et al. 2000). Afterwards the monodisperse aerosol is detected in the CPC (figure 6).

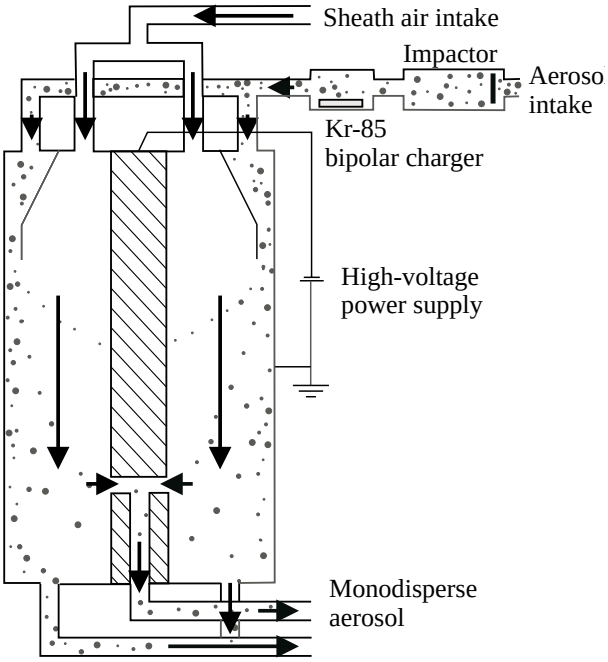

Figure 5:
Schematic of the differential mobility analyser (DMA)

Because particles of diesel exhaust gas are to small for direct optical detection methods they are first enlarged inside the CPC. This is achieved by surface condensation of supersaturated n-butyl alcohol, such that they increase quickly in size (up to about 10 µm). The enlarged particles are counted using laser diode light. The measurement signal used for counting is the scattered light of the single particle (Mayer A. 2001, ACEA 2002).

As alternative to the SMPS technique, the Finnish company Decati Ltd. offers an electrical low pressure impactor (ELPI). For the determination of the particle size distribution samples are taken with a multi-hole probe at the end of the exhaust gas dilution tunnel.

The principal components of the ELPI include a corona discharge, a Berner low pressure impactor, and a series of electrometers. The general scheme of the ELPI is shown in figure 7.

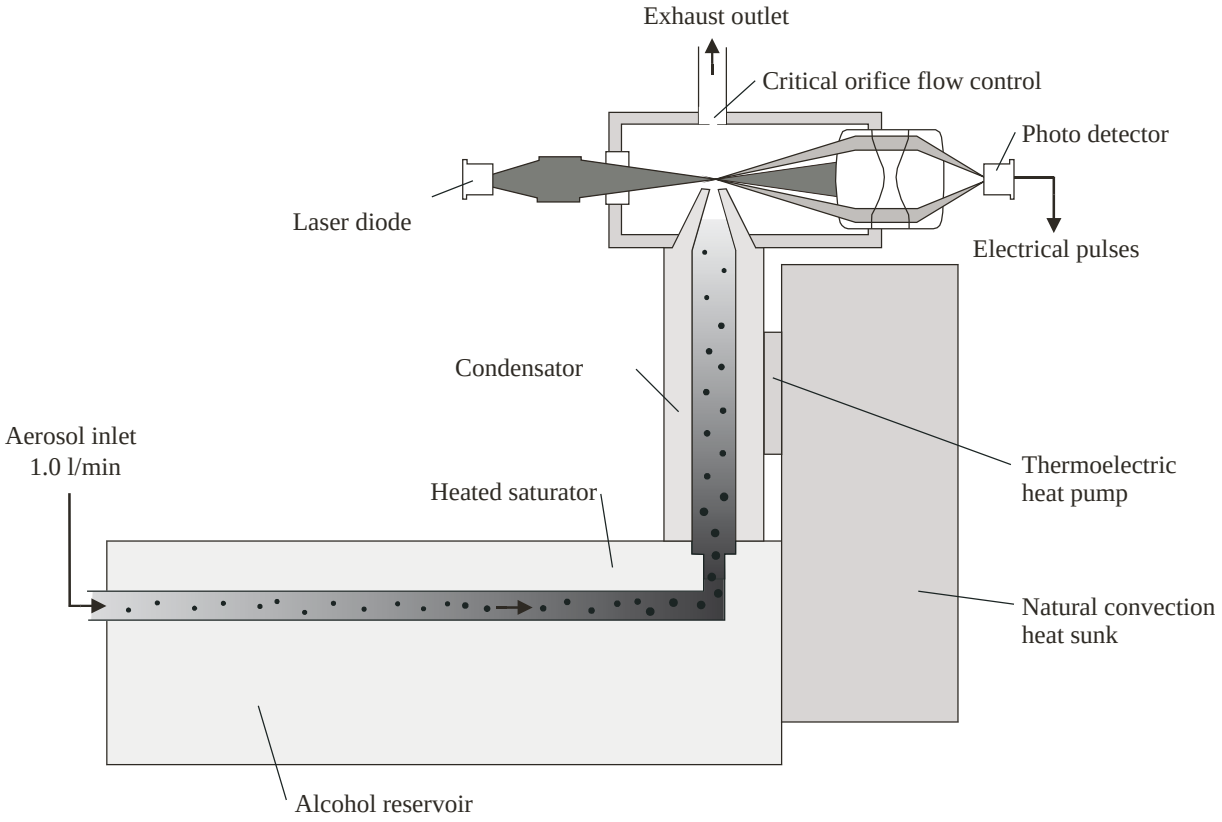

Figure 6:
Schematic of the condensation particle counter (CPC)

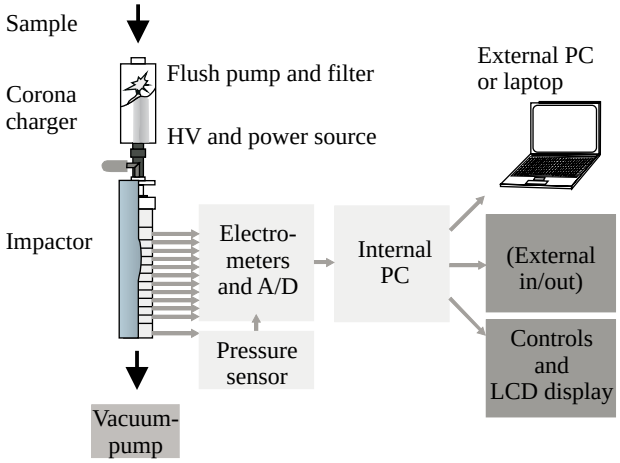

Figure 7:
Schematic of the electrical low pressure impactor (ELPI)

The diode type corona discharge provides unipolar charging of the particles in the sample aerosol. It is coupled with a trap, which has the function to remove ions and small (< 20 nm) particles that can otherwise produce extraneous currents at the electrometers. The amount of charge accepted by the particles increases with their mobility diameters. After being charged the aerosol passes through a thirteen-stage cascade impactor, where the particles are separated in accordance to their aerodynamic diameter. The thirteenth stage removes particles larger than 10 µm. Each of the twelve lower stages is connected to an electrometer that measures the current deposited from particle impaction. The charge on each stage, resulting from particle impaction, is proportional to the number of impacted particles. With a dynamic response of 1 second, the ELPI is able to follow a transient driving cycle (Mayer A. 2001, ACEA, 2002).

Results

PM

PM emissions of the tested fuels are presented in figure 8. The EURO 3 limit of 0.1 g/kWh was met by all four fuels. The lowest emissions were obtained for RME. The highest emissions were determined for DF. The emissions of the two fuels B5Ult and V-Power lay between RME and DF. The value for B5Ult was lower than for V-Power. By use of RME it was possible to obtain reductions in the particle emissions versus DF.

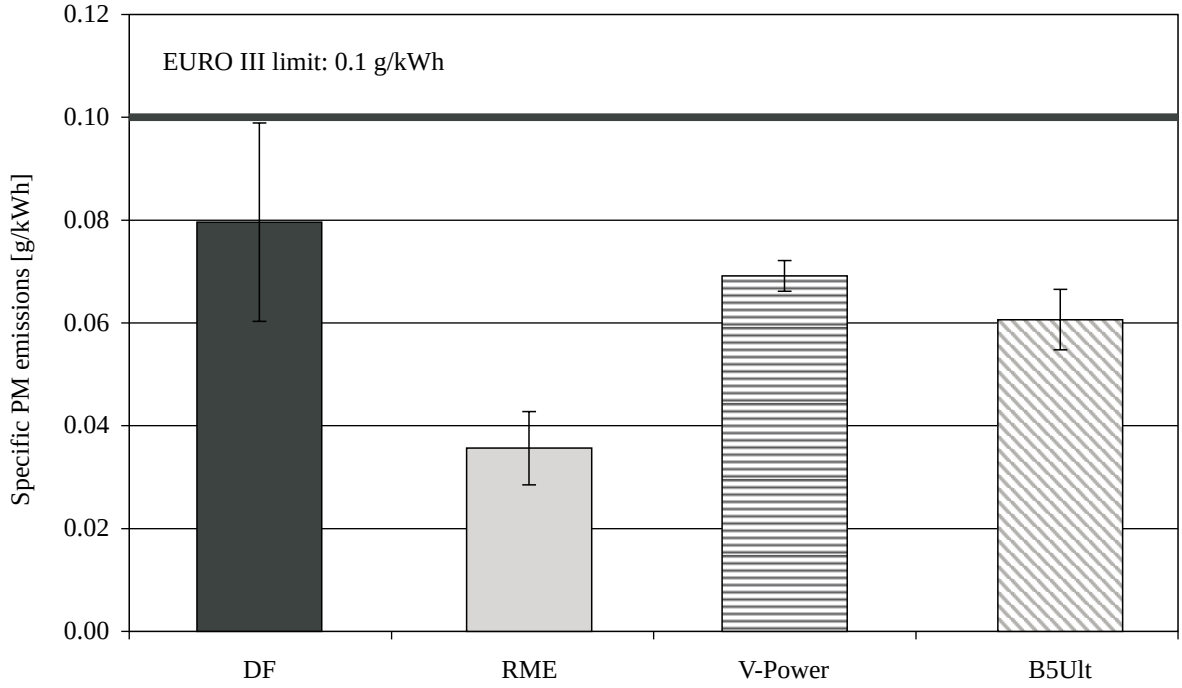

Figure 8:
Specific PM emissions (OM 906 LA, ESC test)

ELPI

The ELPI results are presented in figure 9.

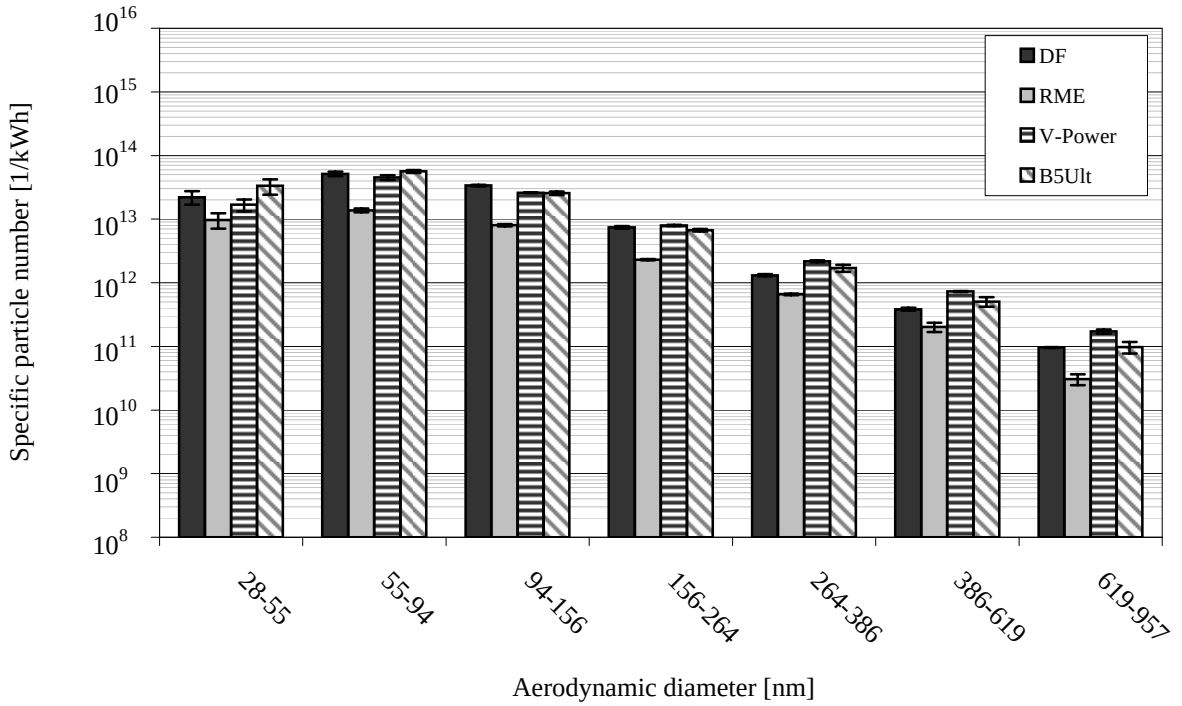

Figure 9:
Specific particle number distribution in the exhaust gas (ELPI, OM 906 LA, ESC test)

RME showed the lowest emissions compared with the other fuels. The other three fuels exhibited a similar emission tendency among themselves over the entire size range. Up to impactor stage that classifies particles with diameters from 156 to 264 nm hardly no differences between V-Power, B5Ult and DF where observed; with exception of the size range from 28 to 55 nm, in which B5Ult showed increasing emissions. As for larger particles the emissions of V-Power and B5Ult were slightly higher versus DF. Starting from the size class range from 156 to 264 nm the highest emissions were detected for V-Power compared with the remaining fuels. In detail, B5Ult showed similar emissions as DF. Advantages concerning the emissions of particles where observed for RME.

SMPS

Analogically to ELPI and PM results, RME led as well to the lowest particulate emissions, when SMPS is used, figure 10.

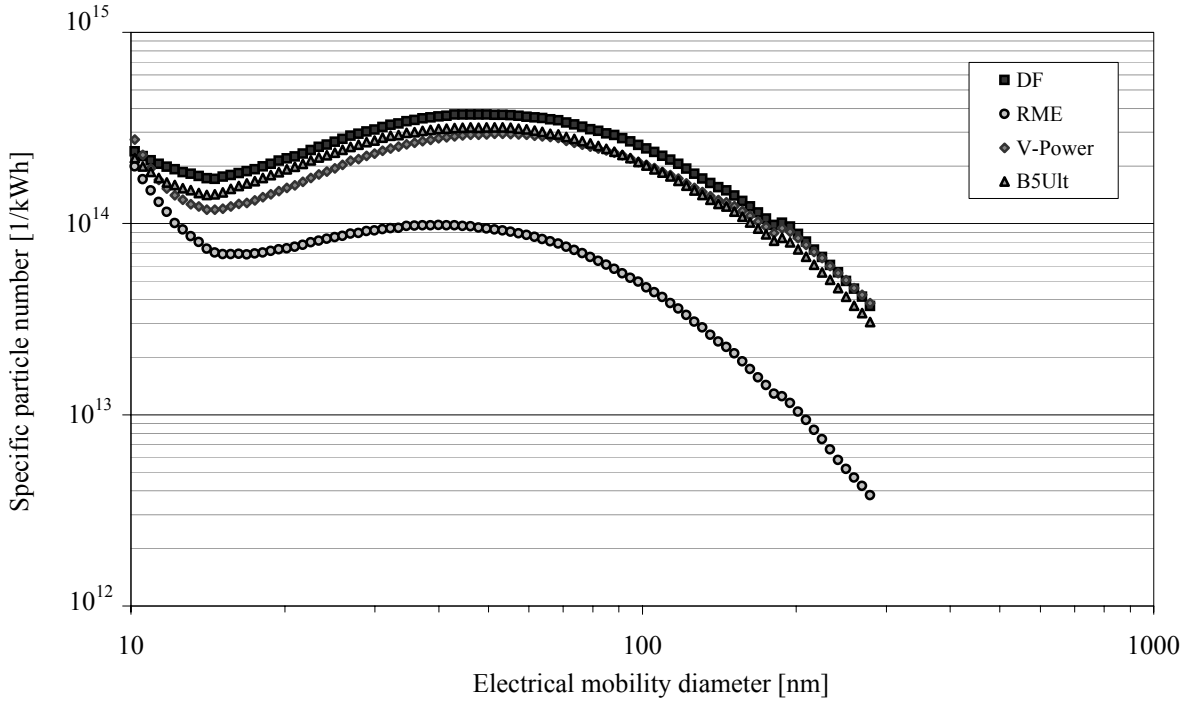

Figure 10:
Specific particle number distribution in exhaust gas (SMPS, OM 906 LA, ESC test)

Besides RME, all diesel fuels showed comparable emissions. However, the particulate emission of reference diesel fuel was slightly increased in comparison with V-Power and B5Ult.

RME demonstrated great potential to obtain a reduction in particle emission.

The determination of the particle size distribution with SMPS shows a bimodal size distribution (figure 1 and 10). In detail, the particle size distribution determined by the SMPS was slightly shifted to smaller values, such that the maximum of the nuclei mode was below 10 nm.

However, in individual modes different distributions were detected. At idle a high emission of ultrafine particles was measured (figure 11). At rated power the accumulation mode predominates. In the range of the accumulation mode the highest particle number emissions were found (figure 12). This leads to the result that – accumulated over the entire ESC test – a bimodal particle size distribution is observed.

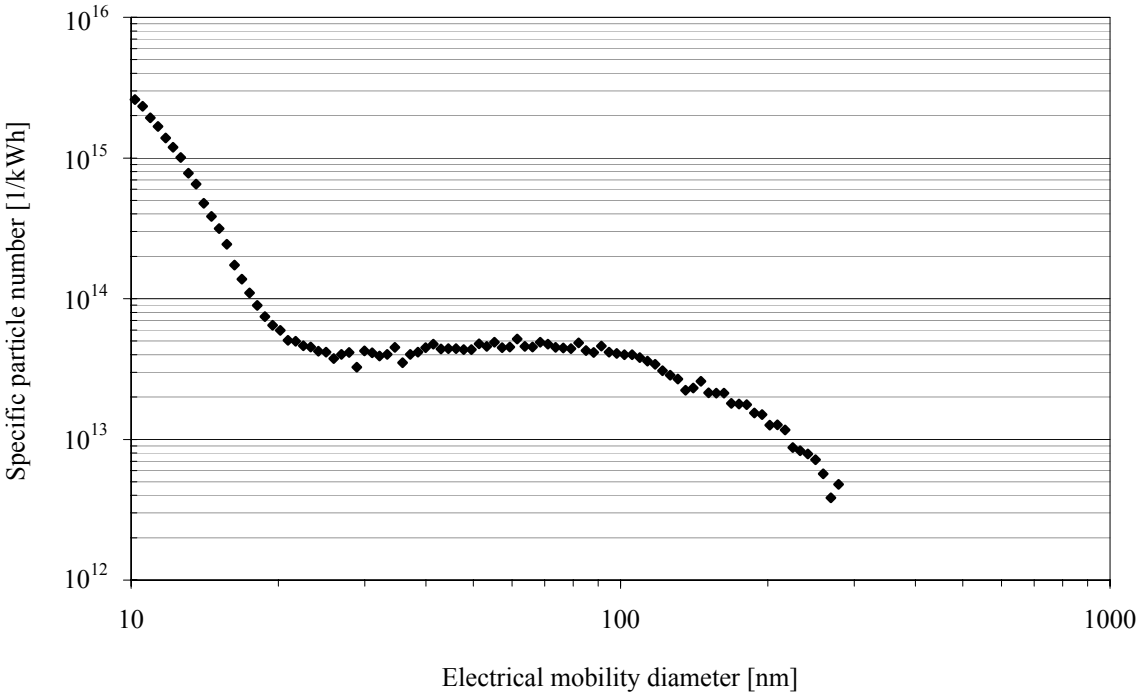

Figure 11:
Specific particle number distribution at idle (SMPS, OM 906 LA, RME)

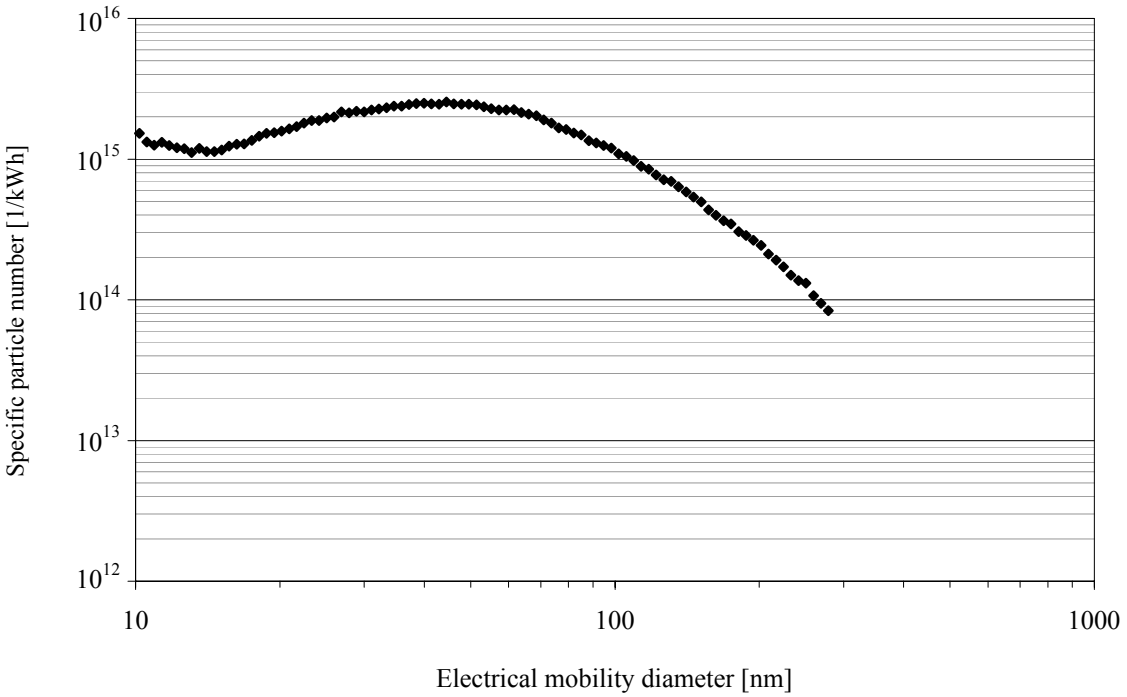

Figure 12:
Specific particle number distribution at rated power (SMPS, OM 906 LA, RME)

Influence of the dilution conditions on the particle size distribution – Temperature effects

The influence of the dilution temperature on the particle size distribution was investigated at the example of RME. Each measurement was fourfold repeated in mode 9 (1800 min^{-1}; 265 Nm) of the ESC test. This test mode was selected, since in this mode both the nuclei mode and the accumulation mode are present and therefore the influence of the temperature on both ranges was expected to be observable quite well.

The influences of the temperature on the particle size distribution are shown in figure 13.

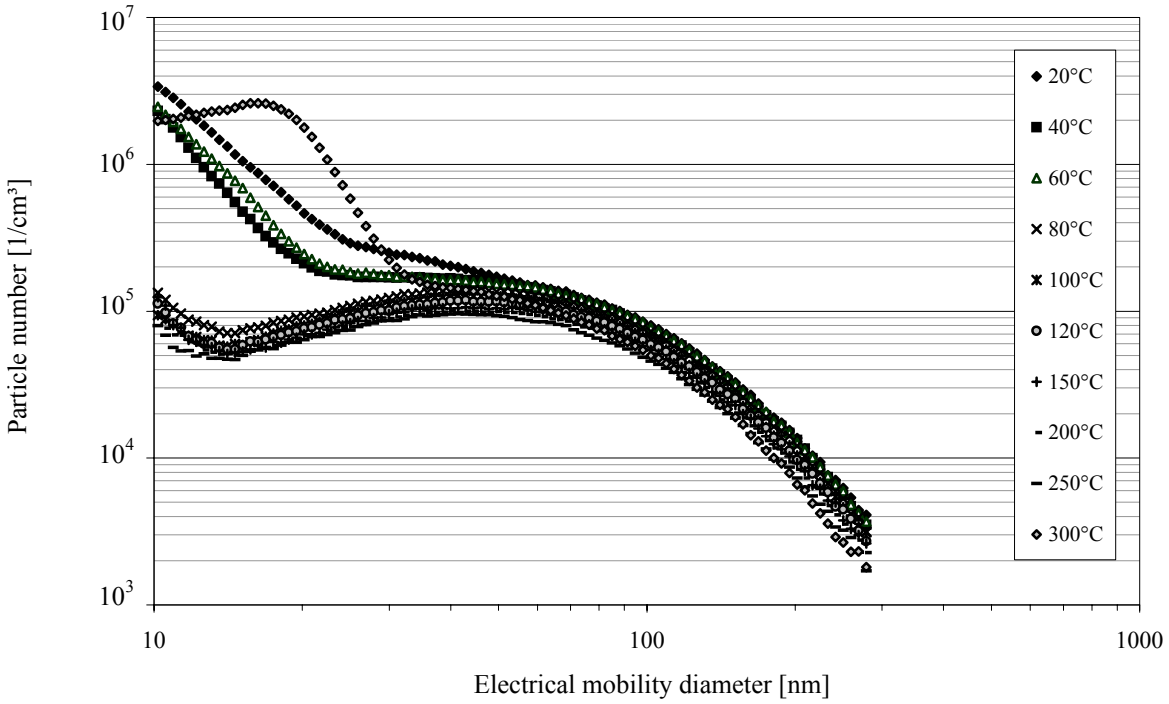

Figure 13:
Particle number distribution at mode 9 (SMPS, OM 906 LA, RME)

In the temperature range from 20 to 60 °C no differences could be observed. The distribution was dominated by particles within the range of 10 nm to 30 nm. With an increase of the temperature up to 80 °C a decrease in number of nuclei particles to one tenth was observed. In the temperature range from 80 to 250 °C the number of particles remained almost constant. At a temperature of 300 °C an increase of particles with a maximum at 17 nm was observed. The influence of the temperatures on the particle number within the size range from 10 to 20 nm has strong effects already at 80 °C. This suggests that particles in this size range may consist mainly of volatile substances. This result is supported by additional investigations of the particle composition. For RME a high quota of soluble organic fraction (SOF) and only a small amount of insoluble fraction (ISF) was found; data not shown. It can be assumed that particles within the range of 10 to 40 nm, which were measured after the hot dilution at temperatures greater than 80 °C, are mainly soot, metallic ash or heavy hydrocarbons (Montajir R. M. et al. 2006).

Summary

Three methods for particle measurement were introduced, such as the legal method for PM emissions and two techniques to detect particle size (ELPI) and particle number distributions (SMPS).

Using these methods diesel engine emissions of a Euro 3 heavy-duty engine were analysed and compared for rapeseed oil methyl ester (RME), reference diesel fuel (DF) and the premium diesel fuels Shell V-Power Diesel® and Aral Ultimate Diesel®. The latter was blended with 5 % RME (B5Ult).

Previous investigations showed higher numbers of particles in the nuclei mode for RME versus DF. Therefore

more detailed information had to be obtained to what extent RME induces the formation of ultrafine particles, which are considered as harmful to human health.

In course of the actual investigations it could be demonstrated that these nuclei particles did not mainly consist of soot, but most probably of unburned fuel. It was shown that the sampling conditions – especially the temperature in the dilution tunnel – had a significant effect on the detected particle size and number distributions. Particularly ultrafine particles in the nuclei mode from 10 to 40 nm decreased when the temperature rose over 80 °C.

RME led to reductions of particle number emissions versus all other fuels, whereas the fossil based diesel fuels DF, B5Ult and V-Power did not vary significantly among each other.

Regarding particulate matter emissions (PM) a reduction by factor two was detected for RME versus DF.

In the result and at the example of the test engine it can be concluded that RME does not lead to higher ultrafine particle emissions than DF, B5Ult and V-Power.

Acknowledgments

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Composition of dust and effects on animals

J. Hartung[1] and M. Saleh[1]

Abstract

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 microorganisms. The dust contains high amounts of proteins and carries gases, odours, microorganisms, and endotoxins. About 85 % of its mass consists of organic material. There are clear differences in the composition of dust originating from types of animals and different housing systems. 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 role of compounds like endotoxins and drug residues (e.g. antibiotics) which can be found in animal house dust in relatively large amounts is not yet sufficiently clear. The aerodynamic diameter of the particles determines how deeply they can penetrate into the respiratory tract. Dust 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. Their impact can be described as mechanical, chemical, infectious, immunosuppressive, allergic, and toxic. Although there is distinct evidence that high dust levels in pig houses reduce production significantly, dust reduction measures are not yet common. It seems necessary to establish scientifically based maximum levels for dust in livestock houses. This would benefit both animals and farmers. For the development of an infectious disease the presence of the respective infectious agent seems to be necessary. Measures to reduce dust formation and increase hygiene in the animal 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. The role of airborne particulates in livestock production needs close further scientific attention.

Keywords: dust, composition, livestock, poultry, bio-aerosol, micro-organisms, endotoxin, animal health

Introduction

There are numerous reports about dust concentrations in animal house air (Takai H. et al. 1998, Hartung J. 1997, Saleh M. 2006). Less information is available on the composition of this dust. Because of the complex nature of animal house dust the term bio-aerosol was introduced (Hirst J. M. 1995, Seedorf J. and Hartung J. 2002). 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} µm. They are widely considered to be principal risk factors for respiratory diseases (Clark S. et al. 1983; Donham K. et al. 1986; Bruce J. M. and Sommer M. 1987). The particulates in animal houses air originate from the feed, the litter, the manure and the animals themselves. These sources determine also the composition of the dust. Dust particles and micro-organisms usually 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 J. 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 S. and Morrison W. D. 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 S. and Morrison W. D. 1988; Perkins S. and Morrison W. D. 1991). These factors can influence the formation, decay, amount, and composition of the airborne particles.

This paper will give an outline of the composition of dust in livestock buildings, cattle, pig, turkey, ducks and laying hens. The potential effects on health and production of farm animals will also be considered.

Aerosols, 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

[1] Institute for Animal Hygiene, Animal Welfare and Behaviour of Farm Animals, University of Veterinary Medicine Hannover, Hannover, Germany

approximately 10^2 µm. 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 H. 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 G. A. et al. 1986). `Dusts' are dispersed particles of solid matter in gases, which arise during mechanical processes or have been stirred up. They belong to the aerosols together with smoke and fog (Henschler D. 1990). Dust may cover a wide range of sizes and can be airborne or settled (De Boer S. and Morrison W. D. 1988). It must be seen as a significant atmospheric contaminant and should no longer be perceived as a mere nuisance (Honey L. F. and McQuitty J. B. 1976).

Origin and composition of animal house dust

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, feed 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 J. 1986). A small amount of dust also comes from the air intake into the house (Dawson J. R. 1990). A compound which is regularly found in dust samples from all animal houses is endotoxin. In addition, dust particles can carry residues of drug applications in an animal house.

Endotoxins in sedimentation dust

Dust samples were taken by the help of standardized sedimentation plates in various animal houses with different species. The results are summarized in Table 1.

Table 1:

Mean, min/max values and standard deviation (s) of endotoxin concentrations (EU/g) in sedimentation dust from animal houses of different species. Cattle a = with straw, cattle b = without straw

	Broiler	VOL	AKA	AKB	Turkey	Ducks	Pig	Cattle a	Cattle b
n	8	8	9	9	9	3	10	3	3
Mean (EU/g)	736	1169	1316	1628	1612	855	1143	669	447
s	360	341	535	753	1128	84	809	471	334
Maximum (EU/g)	1178	1740	2240	3295	4131	946	2624	1196	796
Minimum (EU/g)	376	522	518	942	339	782	480	289	131

n*= Number of monthly samples. Vol = aviary. AKA = furnished cage type Aviplus. AKB = furnished cage type Eurovent.

The measurement of dust 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 dust 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 dust fractions can be described as nose-pharynx-larynx dust, tracheobronchial dust and alveolar dust (Henschler D. 1990). Additionally, the nature of the particles and the compounds which they are carrying determine the health effects.

The highest single concentration was found with 4131 x 10^3 EU/g in the dust from a turkey barn. Taking the arithmetic mean, the highest concentrations were found in AKB (1.6 mio EU/g) followed by turkey, AKA, VOL and pig. The concentrations of endotoxin in the dust of broiler and duck barns were about 50 % lower. The lowest concentrations were found in the cattle house without straw. The relative high endotoxin concentrations in the dust from AKA and AKB may have been influenced by the position of the sedimentation plates which were placed in these systems on the level of the manure belts.

Material composition of the dust

From an earlier investigation of the composition of sedimentation dust from a piggery and a poultry house, it is known that up to 85 % of the dust consists of organic matter. Table 2 shows the results. The crude protein content of the dust is noticeably 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. Dust from stables was found to contain over 26 % protein (Zeitler M. 1988).

In a recent study (Saleh M. 2006) the protein content in the dust from pig and poultry buildings was even higher. Table 3 summarizes the results of the analysis of the dust according to protein, fat, fiber, nitrogen free components and ash.

Table 2:
Composition (%) of feed and deposited dust from a piggery and from a poultry house (after Hartung J. 1983; Aengst C. 1984)

Component	Pig house Deposited dust	Pig house Feed	Poultry house Deposited dust
Dry matter	78	88	89
Crude protein	24	19	50
Crude fat	4	4	10
Crude fibre	3	5	-
Crude ash	15	5	-

The dust from poultry houses contains the highest amounts of protein. This is caused firstly by the relatively high protein content in the feed which is usually between 20 and 25 % (Kamphues J. et al. 2004). The other proportion of up to 45 % comes probably from feathers and claw abrasion. Also in the pig house dust a percentage of about 20 % seems to come from the skin and the hairs of the animals. The relatively high amounts of fat in the dust from the duck house can be explained by the behaviour of the duck to use a self produced fatty substance for preening and protecting the feathers. The high fiber content in the turkey house dust may have been influnced by the hay baskets which were offered to the turkeys as a mean to prevent feather pecking and cannibalism.

Amounts of microorganisms in dust

Animal house dust is an important carrier of bacteria and fungi. Table 4 gives an overview of the amounts of total bacteria count, E. coli, staphylococci, streptococci and fungi in the sedimentation dust from animal houses of 6 different species.

Table 3:
Mean (a) and standard deviation (s) of dust analysis from different animal houses in percent (%) of dust mass. T.S = dry matter. R.A = crude ash. R.P. = crude protein. R.Fa. = crude fiber. N.F.E. = nitrogen free substances

		T.S. %	R.A. %	R.P. %	R.F. %	R. Fa. %	N.F.E. %	
Broiler	a	90.8	9	70.6	3.8	4.7	2.6	N = 8
	s	0.8	1.4	3.7	0.5	2.5	2.3	
Aviary	a	88.8	13	62.3	3.6	6.2	3.6	N = 11
	s	1.3	1.5	7.7	1.9	3.7	5	
Furnished cage	a	90.1	11.7	63.9	6.4	3.7	4.3	N = 11
	s	0.6	1.5	4.6	1.5	2	3.2	
Ducks	a	89.4	8.6	58.6	8.4	3.1	10.6	N = 6
	s	2.3	1.2	3.8	0.8	1.7	2.8	
Turkeys	a	89.8	9	53.8	3.4	11.2	12.4	N = 11
	s	1.6	1.9	6	1	4.4	5	
Pigs	a	90.2	14.5	38.4	3.6	1.3	32.5	N = 13
	s	1	1.2	2.9	0.9	0.7	3.6	
Cattle	a	85.8	18.8	29.7	6.6	6.6	24.1	N = 6
	S	5,4	3.8	4.3	1.7	1.7	3.1	

All dust samples contain relatively little water. In poultry and pig dust the dry matter content reaches about 90 %. The dust from the cattle barn showed a humidity of about 15 %. This value is slightly above the threshold of bacterial growth which is around 14 % moisture in dry meal feed (Kamphues J. et al. 2004). The relative high content of ash in the dust from the cattle house is probably due to sandy feedstuff. The very low values in the moskovy duck house are probably caused by the plastic flooring.

The highest concentrations are observed in broiler barns followed by duck, turkey, laying hen, pig and cattle. This reflects the common experience that the highest concentrations of micro-organisms are found in poultry houses. There are clear differences between keeping systems of one species. In the aviary (VOL) distinctly higher amounts of bacteria were found compared to furnished cage systems.

Table 4:
Mean concentrations (a) of typical micro-organisms in sedimentation dust from animal houses of different species. Standard deviation (s)

		Total count cfu/g x 10^6	E. coli cfu/g x 10^3	Staph. cfu/g x 10^6	Strep. cfu/g x 10^6	fungi cfu/g x 10^5
Laying hens						
Aviary	a	1741	57	1168	5	4
	s	1146	29	676	4	6
Laying hens	a	200	398	124	8	12
Aviplus (AKA)	s	82	352	55	3	12
Laying hens						
Eurovent (AKB)	a	408	499	342	12	4
	s	268	373	228	2	5
Broiler						
	a	7772	285	6326	22	289
	s	3990	177	2933	24	129
Turkeys	a	3554	1813	2915	93	71
	s	1938	1304	1866	31	53
Cattle a	a	91.4	1651.6	64.7	1.3	6.3
	s	79.9	1497	59.9	0.6	52
Cattle b	a	85.7	1574	63	0.9	5.7
	s	8.1	109	1.6	0.5	8
Moscovy duck						
	a	5782	129	1764	224	3
	s	3644	150	1956	193	6
Pig						
	a	88	90	26	15	2
	s	44	96	17	10	4

The dust from livestock buildings contains a variety of further compounds which are potentially hazardous agents (Donham K. J. 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 J. 1985).

Further compounds which are associated with animal house dust are drugs. In a recent study Hamscher G. et al. (2003) were able to show that pig house dust has an excellent “memory” for all types of antibiotics. Up to 12 mg/kg dust of antibiotic residues were analyzed. It was possible to identify different classes of antibiotics in the dust even 15 years after its application.

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. Particulates can have effects which may be described as mechanical, infectious, immunosuppressive, allergic or toxic. Table 5 summarizes the possible effects of airborne dust, microorganisms and gases on animal health.

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 R. R. 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 J. H. and Mark D. 1981; Henschler D. 1990).

A deciding factor in the depth of penetration is the aerodynamic diameter. At diameters of 4 to 5 µm 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 G. A. 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 weaner house for the first 20 days after weaning, 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.

Table 5:
Possible influences of dust and microorganism levels on animal health (after Zeitler M. 1988)

Factor	Effect on the animal
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

High levels of aerial pollutants can distinctly influence the health status of pig herds. In a comparison of 44 fattening pig herds with (+AR n = 15) and without (-AR n = 29) clinically recognized atrophic rhinitis (AR) was shown that increased levels of dust, bacteria, endotoxins, and ammonia were associated with a higher incidence of AR (Baekbo P. 1990). When comparing these results with a proposal for an exposure threshold given by Donham K. J. (1989), it is shown that respiratory disease may be absent even under unfavourable concentrations of some of the factors.

A lot of attention has been given to endotoxins which seem to be implicated in the pathogenesis of hypersensitive pneumonias in humans (Thelin A. et al. 1984). Apart from their allergic potential, they also affect the immune system (Rylander R. 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 F. J. 1982). The role of endotoxins in respiratory diseases of 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 (Rylander R. et al. 1989).

Dust 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. High dust burden can distinctly influence the health of animals kept in confined buildings. For the development of an infectious disease the presence of the respective infectious agent seems to be necessary. Measures to reduce dust formation and increase hygiene in the animal houses should be given more sustained application in practice. It seems necessary to establish scientifically based maximum levels for dust in livestock houses. This would be in the interests of workers, of animal welfare, and of the wider environment. The role of airborne particulates in livestock production needs close further scientific attention.

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Comparison of odorants in room-air and in headspace of sediment dust collected in swine buildings

H. Takai[1], P. J. Dahl[1], M. Maahn[1], J. O. Johnsen[1], and P. Kai[1]

Abstract

This paper presents preliminary results from a study aimed at comparing odorants in room-air and odorants in static headspace of sediment dust samples collected in swine buildings. The study included the samples collected during winter and summer surveys at four different swine farms, each equipped with gestation, farrowing, rearing, and finishing buildings.

The room-air odorants in different swine buildings were sampled using solid-phase microextraction (SPME) fibres and their relative concentrations were analyzed using gas chromatography and mass spectrometry (GC/MS). On each sampling occasion, two SPME samples were taken and a sediment dust sample was collected from the surfaces of fixtures in the room. About 5 g of the sediment dust sample was packed in a ∅ 38 mm stainless steel pipe, whose headspace was connected with a closed air circulation pipe. In a climate chamber, the whole setup was kept at 30 °C for 30 min to achieve the equilibrium stage, then, the dust-headspace odorants were sampled using SPME. Then, their relative concentrations were analyzed by GC/MS. The odorants included in the present study are: trimethyl amine (TMA), dimethyl sulphide (DMS), butanoic acid (BA), 3-methyl-butanoic acid (3MBA), 4-methyl-pentanoic acid (4MPA), benzyl alcohol (BAL), indole (IND), and 3-methyl-indole (3MIND). These 8 odorants are called technical key odorants (TKOs).

As overall, TKO concentrations in room-air and headspaces are significantly correlated; but a relatively large spreading was also observed. Further study on interaction between TKOs in room air and dust particles is recommended.

Keywords: odour, swine building, dust headspace, SPME

Introduction

More than 300 different odorous compounds have been identified in livestock buildings (Tanaka H. 1988, Schiffman S. S. et al. 2001). Tanaka H. (1988) reported that ammonia, sulphides, volatile fatty acids, amines, indoles, phenols, ketones, aldehydes and mercaptans are compounds/compound groups of special importance in connection with offensive odours from animal husbandry. Many of these odorants are generated in slurry under anaerobic conditions. The profile of the odorant concentrations in room air is affected by different conditions, such as feeding, management of livestock manure, hygiene and ventilation methods, and the rate of air exchange (Jacobson L. D. et al. 2000, Janni K. et al. 2001). Hartung J. (1985) showed that dust from swine confinement buildings contains VFA, phenols, indoles and scatole. The airborne dust originated from manure contains these compounds. The dust particles interact with the surrounding air and may exchange the odorous compounds by means of ad-, ab- and desorption processes. Hammond E. G. et al. (1981) estimated that the concentration of butyric acid and p-cresol will be about 4×10^7 greater in a dust particle than in an equal volume of air. Reynolds S. J. et al. (1998) estimated that a significant proportion (15 - 23%) of airborne ammonia in enclosed livestock facilities is associated with particles. Takai H. et al. (2002) reported that ammonia contents in inhalable dust collected in dairy, poultry and farrowing houses ranged from 1-6 µg per mg of dust, i.e. 1,000 to 6,000 ppm on a weight basis, while about 7 µg NH_3 per mg of dust, i.e. 7,000 ppm, was found in respirable dust.

Ammonia, hydrogen sulphide and other odorous compounds in the respirable fraction of inhaled dust particles may reach the lower parts of the respiratory tract, i.e. the bronchi and the alveoli, and irritate the organs. If this hypothesis is true, the chemical compounds absorbed in dust particles plays an important role in the development of respiratory diseases in farmers' lungs. The deposition of dust particles in the nose may result in high local concentrations of odours at and in the mucosa in the *regio olfactoria*, which will affect the perception of odour.

To understand the synergistic effects of gases and aerosols on farmers' health and malodour problems improved knowledge on relation between odorant concentrations in dust particles and in room air is desired.

[1] Aarhus University, Department of Agricultural Engineering, Research Centre Bygholm, Horsens, Denmark

The present study is aimed at comparing the odorants in the room-air and in the headspace of the sediment dust samples collected in swine buildings.

Method and materials

The study have included samples collected during winter and summer surveys at four different swine farms, each equipped with gestation, farrowing, rearing, and finishing buildings. Two surveys were carried out in the winter of 2003–2004 and two in the summer of 2004.

The odorants included in the present study are: trimethyl amine (TMA), dimethyl sulfide (DMS), butanoic acid (BA), 3-methyl-butanoic acid (3MBA), 4-methyl-pentanoic acid (4MPA), benzyl alcohol (BAL), indole (IND), and 3-methyl-indole (3MIND). These 8 odorants are called technical key odorants (TKOs) and were selected based on the results from earlier study (Takai H. et al. 2007), who applied principal variable analysis to identify TKOs. The study showed that these 8 TKOs could explain more than 90% of the variation found in a data set of 18 room-air odorants. The physical properties and odor threshold levels of TKOs found in the literatures are shown in table 1.

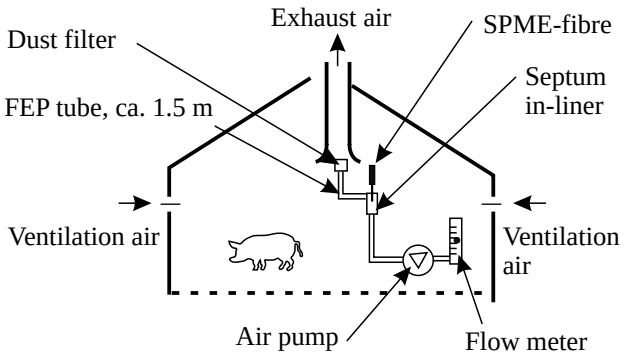

Figure 1:
Odorant sampling in a swine building by means of SPME

About 5 g of the sediment dust sample was packed in a ∅ 38 mm stainless steel pipe, of which the headspace air was circulated in a closed system consisting of FEP tubes, a septum in-liner and a stainless steel air pump (AFC 123 personal air sampler; BGI Inc., USA). In a climate chamber, the setup was kept at 30°C for 30 min to achieve the equilibrium stage, then, the dust-headspace odorants were sampled by using SPME, figure 2. Then, their relative concentrations were analyzed by GC/MS.

Table 1:
Physical properties and odor threshold levels of the technical key odorants (TKOs) included in the present study (Syracuse Research Corporation's, 2007)

Name	CAS #	Mol weight m	Solubility H_2O (25°C)	Boiling point	Vapor Pressure (25°C)	Henry's constant k_h	Dis-sociation constant	Odour threshold[2)]
		g mol^{-1}	g l^{-1}	(°C)	mm Hg	atm l mol^{-1}	pK_a	µg m^{-3}
Trimethyl amine (TMA)[1)]	75-50-3	59.11	890.0	2.8	1.61E+03	1.04E-01	9.80	5.9
Dimethyl sulphide (DMS)	75-18-3	62.13	22.0	37.3	5.02E+02	1.61E+00		5.9
n-Butyric acid (BA)	107-92-6	88.11	60.0	164.1	1.65E+00	5.35E-04	4.83	14.5
3-Methyl butanoic acid (3MBA)	503-74-2	102.10	40.7	176.5	4.40E-01	8.33E-04	4.77	10.5
4-Methyl pentanoic acid (4MPA)	646-07-1	116.16	5.3	200.5	4.45E-01	1.70E-03	4.84 15.40	75.9 24,500.0
Benzyl alcohol (BAL)	100-51-6	108.14	42.9	205.3	9.40E-02	3.37E-04	0	0
Indole (IND)	120-72-9	117.20	3.6	253.0	1.22E-02	5.28E-04	-2.40	0.2
3-Methyl-1H-indole (3MIND)	83-34-1	131.10	0.5	265.0	5.55E-03	2.13E-03	-3.40	3.1

[1)] As a solution in water ~50%
[2)] Schiffmann S. S. et al. (2001)

The room-air odorants in different swine buildings were sampled using solid-phase microextraction (SPME) fibres (figure 1), and their relative concentrations were analyzed using gas chromatography and mass spectrometry (GC/MS). On each sampling occasion, two SPME samples were collected in parallel and a sediment dust sample was collected from the surfaces of fixtures in the room. The sediment dust samples were kept in airtight blue cap glass bottles (100 ml) with PTFE sealing in a freezer at -15°C until analysis within about 120 days.

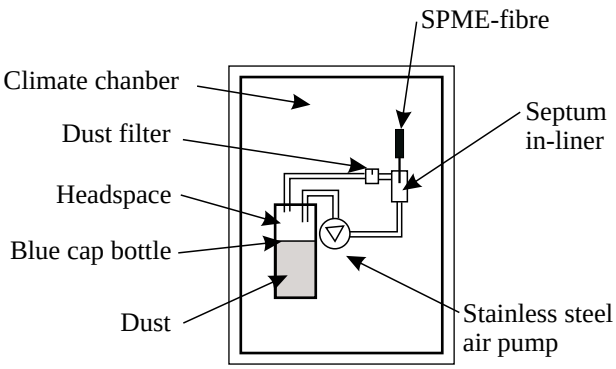

Figure 2:
Sampling of odorants of dust headspace air at different temperatures

Solid-phase microextraction for determining the relative odorant concentrations:

SPME is a qualitative analysis technique. To conduct *quantitative* analysis, a calibration method would have to be developed, which was not possible in the present study. It was assumed that the amount of molecules of an odorant sampled using SPME would indicate its relative concentration. This assumption apparently works for comparing odors composed of similar chemical compositions, e.g., odors in and from swine buildings. To improve the comparability of the relative odorant concentrations in different swine buildings, a standard procedure for SPME sampling was used, as follows. SPME fibers coated with 50/30 µm Divinylbenzene/Carboxen/Polydimethylsiloxane (Supelco, Bellefonte, Pennsylvania, USA**)** were inserted into a Polytetrafluorethylene (PTFE) septum injector (Omnifit, Cambridge, England), which was inserted between a dust filter (25 mm of 0.2-µm PTFE membrane syringe filter, Whatman, Brentford, Middlesex, UK) and an air pump with a suction sampling rate of 0.001 m^3 min^{-1} (fig. 1). The septum injector ensured that all SPME fibers used in the surveys were exposed to the air samples under the same air flow conditions. The sampling period was 10 min, equivalent to a sampled air volume of 0.01 m^3. This sampling period was chosen on the basis of pre-surveys indicating that an SPME fiber has a much greater absorption capacity than the amount of odorant molecules that can be absorbed in swine buildings within the applied sampling period.

The amounts of different odorant molecules sampled by means of SPME were analyzed using a Varian 3800 gas chromatograph (GC) coupled to a Varian Saturn 2000 ion trap mass spectrometer (MS) (Varian; Palo Alto, CA., USA). The GC was equipped with a special SPME liner (Varian) and a Varian VF-1ms Factor Four non-polar capillary column (60 m × 0.32 mm × 1.0 µm). Helium Alphagaz 2 (Air Liquide Danmark, Ballerup, Denmark) was used as carrier gas at 2.5 mL min^{-1} (constant pressure at 1×10^5 N m^{-2}). The SPME fiber was inserted directly into the injector for five min at 250°C. The oven temperature program was as follows: 4 min at 40°C , a ramp to 150°C at 5°C/min, followed by a ramp to 220°C at 10°C/min, and 7 min at 220°C. The MS was used in the electron impact mode at 70 eV and with a scanning range of 30–150 m/z. The temperatures were as follows: ion trap, 175°C; manifold, 110°C; and transfer line, 170°C.

It was estimated that the detection limit (DL) of the applied GC/MS analysis was approximately 500 counts. In the case of a count less than 500, a dummy value of DL/2 = 250 (which is almost zero compared to the GC/MS counts representing the concentrations typically found in the samples) was inserted to allow log transformation before further statistical analysis. Log transformation was performed mainly because the distributions of the original data were skewed, with long tails to the right.

Results and discussions

Technical parameters, which assumed to have influence on odour and dust emissions in the swine buildings, were determined at the sampling occasions. The maximum, minimum and average values of the technical parameters are shown in table 2a and 2b. The final datasets subjected to the statistical analyses were derived from 6, 14, 11 and 13 averaged values of duplicate measurements of room-air odorant and from the corresponding 6, 14, 11 and 13 measurements of odorants in the headspace of the sediment dust samples collected in gestation, farrowing, rearing and finishing buildings, respectively. The reason for the small observation number for gestation building is that the surfaces in the building were often too wet to be able to collect a satisfactory amount of sediment dust. Table 3 shows the number of observations, where the TKO concentrations were higher than DL. As seen in the table, DMS concentrations have often been lower than DL especially in the gestation and farrowing buildings. Only 6 out of 44 measurements on DMS in dust headspaces showed higher concentrations than DL. All 6 measurements on 4MPA in room-air of gestation buildings were lower than DL. While 4MPA was observed more frequently in the room-air of the other buildings types . Dust headspaces for gestation, farrowing and rearing buildings often showed 4MPA concentration lower than DL, whereas, the 4MPA was more common compound in the dust headspaces of finishing buildings.

Table 2a:
Ranges and averages of some technical parameters determined in connection with sampling of odorants in the swine house

	Gestation			Farrowing		
	Max.	Min.	Ave.	Max.	Min.	Ave.
Room:						
Rom volume, m^3	1122	468	734	870	92	644
No. of pens, $room^{-1}$	180	78	121	75	16	55
Area of slatted floor, %	100	50	67	33	33	33
Animals:						
Body weight, kg pig^{-1}	250	150	192	250	200	225
Heat production, W $animal^{-1}$	490	393	434	473	426	449
Stocking density:						
No. of animals, $room^{-1}$	189	68	114	75	16	49
Do., m^{-2}	0.4	0.3	0.4	0.4	0.1	0.2
Boddy weight, kg m^{-3}	42.1	20.5	29.3	43.5	10.0	18.8
Do., kg m^{-2}	105.2	51.3	72.7	100.0	28.7	47.2
Indoor climate:						
Ventilation rate, $m^3\ h^{-1}\ hpu^{-1}$	247	86	154	336	76	229
Do, $m^3\ h^{-1}\ room^{-1}$	11810	2869	7234	10095	965	5030
Air exchange rate, h^{-1}	14	6	10	16	2	8
Temperature	24	18	21	27	18	23
Relative humidity	76	61	69	73	47	63
NH_3 concentration, ppm	18	6	12	14	3	6
CO_2 concentration, ppm	2500	1100	1733	2800	900	1371
Room cleanliness:						
1-5: Dry floor - Wet floor	4	2	2.7	3	1	1.5
1-5: Not dusty - Very dusty	3	1	2.0	4	1	2.5

Table 2b:
Ranges and average of some technical parameters determined in connection with sampling of odorants in the swine houses

	Rearing			Finishing		
	Max.	Min.	Ave.	Max.	Min.	Ave.
Room:						
Rom volume, m^3	427	104	224	1008	330	703
No. of pens, $room^{-1}$	8	5	7	28	9	16
Area of slatted floor, %	100	33	51	100	100	100
Animals:						
Body weight, kg pig^{-1}	30	12	20	80	50	68
Heat production, W $animal^{-1}$	130	73	101	217	174	202
Stocking density:						
No. of animals, $room^{-1}$	300	125	187	560	75	267
Do., m^{-2}	3.8	1.1	2.4	1.5	0.4	1.1
Boddy weight, kg m^{-3}	46.2	6.6	21.0	38.7	8.2	25.1
Do., kg m^{-2}	92.3	16.4	49.8	109.1	32.8	75.8
Indoor climate:						
Ventilation rate, $m^3\ h^{-1}\ hpu^{-1}$	1233	76	299	529	59	203
Do, $m^3\ h^{-1}\ room^{-1}$	14314	1190	4876	57036	2055	12139
Air exchange rate, h^{-1}	95	5	28	66	4	17
Temperature	24	19	22	26	17	21
Relative humidity	77	51	65	83	52	67
NH_3 concentration, ppm	8	1	3	24	6	14
CO_2 concentration, ppm	2800	500	1545	3500	700	1638
Room cleanliness:						
1-5: Dry floor - Wet floor	4	1	2.5	4	1	2.3
1-5: Not dusty - Very dusty	3	1	2.5	4	2	2.8

Figure 3 shows the correlation between odorant concentrations in room-air and dust headspace. The trend line in the figure shows the overall trend of the 8 odorants. Although the overall correlation is significant some of TKOs show relatively large spreading. To explore a possible effect of building type on the interaction between room-air and dust particles, correlation coefficients between relative TKO concentrations in the room-air and in the dust headspaces were determined for different building types. The results are shown in table 4. None of the correlation coefficients for the gestation building were significant. One reason for this might be the small number of observations, n=6. Farrowing buildings showed that TMA, BA and 3MBA concentrations in room-air and dust headspace were significantly correlated. TMA, DMS, BA and 3MBA concentrations in rearing buildings were significantly correlated. While in the finishing buildings, 5 TKOs, i.e. TMA, BA, 3MBA, IND and 3MIND concentrations in room-air and dust headspace were significantly correlated.

Conclusions

- Relative concentrations of 8 TKOs in room-air and dust headspaces samples from gestation, farrowing, rearing and finishing buildings have been determined and their correlations are studied.
- As overall, TKO concentrations in room-air and headspaces were significantly correlated; but relatively large spreading was observed.
- Further study on the interaction between TKOs in room-air and dust particles is recommended.

Table 3:
Number of TKO concentration data higher than detection limit for room-air and dust head spaces for different building types.

Type of buildings	No. of observations	Trimethyle-amine (TMA)	Dimethyl-sulfide (DMS)	Butanoic-acid (BA)	3-methyl-butanoic-acid (3MBA)	4-methyl-pentanoic-acid (4MPA)	Benzyl-alcohol (BAL)	Indole (IND)	3-methyl-indole (3MIND)
	Room-air								
Gestation	6	6	2	6	4	0	5	6	6
Farrowing	14	12	2	14	14	7	12	14	14
Rearing	11	10	6	11	11	10	10	11	11
Finishing	13	12	8	13	13	7	12	13	13
	Dust headspace								
Gestation	6	6	1	6	5	2	6	6	6
Farrowing	14	14	1	14	14	3	13	14	14
Rearing	11	11	2	11	11	6	10	11	11
Finishing	12	13	2	13	13	11	12	13	13

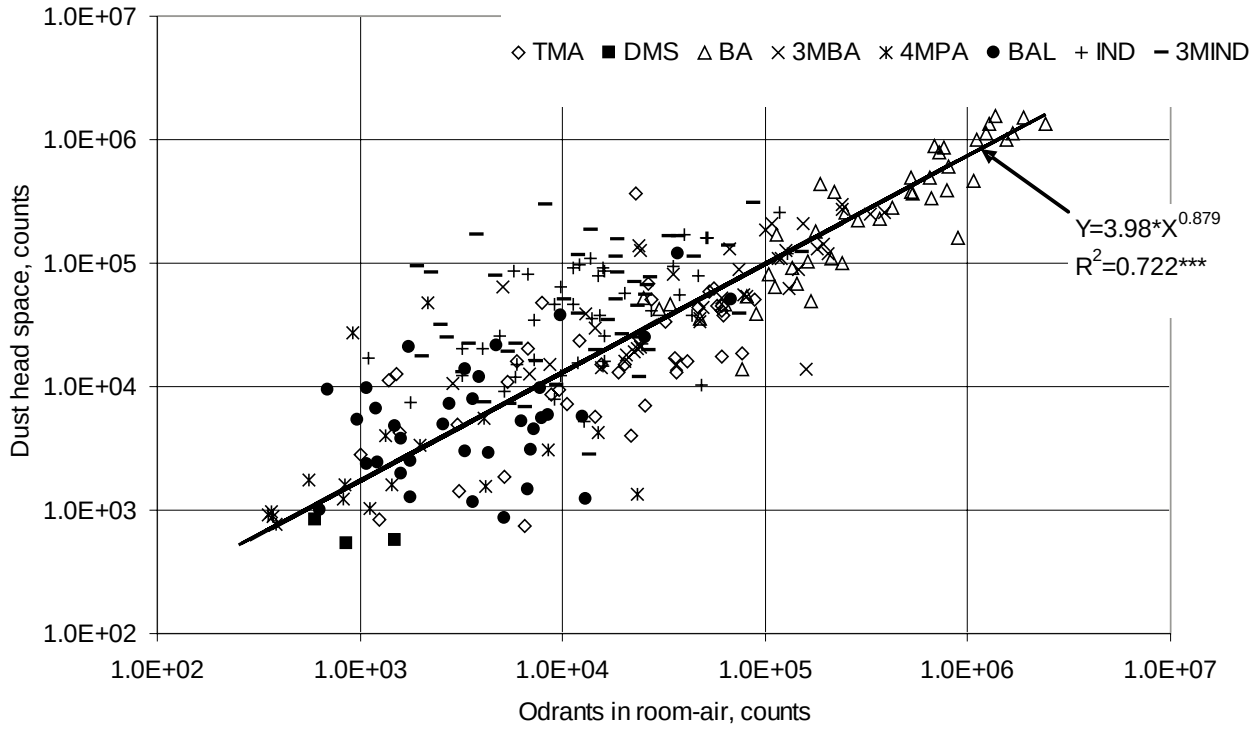

Figure 3:
Correlation between relative odorant concentrations in room-air (X-axis) and dust headspace (Y-axis)

Table 4:
Correlation coefficients between relative odorant concentrations (GC/MS counts) in the room air and in the dust headspace

Type of buildings		Trimethyle-amine (TMA)	Dimethyl-sulfide (DMS)	Butanoic-acid (BA)	3-methyl-butanoic-acid (3MBA)	4-methyl-pentanoic-acid (4MPA)	Benzyl-alcohol (BAL)	Indole (IND)	3-methyl-indole (3MIND
Gestation	r	0.580	-0.299	-0.103	-0.327	-	0.190	0.586	0.450
	P-value	0.132	0.473	0.809	0.430	-	0.653	0.127	0.263
Farrowing	r	0.681	-0.104	0.815	0.681	0.124	0.374	0.141	-0.030
	P-value	0.004	0.701	0.000	0.004	0.648	0.154	0.601	0.912
Rearing	r	0.611	0.678	0.874	0.739	0.397	0.538	0.511	0.305
	P-value	0.027	0.011	0.000	0.004	0.180	0.058	0.074	0.310
Finishing	r	0.663	-0.226	0.788	0.650	0.266	0.231	0.531	0.558
	P-value	0.007	0.418	0.000	0.009	0.338	0.407	0.042	0.031

Aknowledgement

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Abatement, control and regulation of emissions and ambient concentrations of odour and allergens from livestock farming in the Nordic countries

P. V. Madsen[1], O. Hertel[1], T. Sigsgaard[2] J. Bønløkke[2], and K. Puputti[3]

Abstract

A survey on current practise and ongoing policy regarding abatement, control and regulation of emissions and ambient concentrations of odour and allergens from livestock farming in the Nordic countries is planned to form the basis for a common Nordic strategy in this area. Such a strategy would be an important element in reducing the number of people in the Nordic Countries exposed to odour and/or allergens as well as to other livestock related compounds health hazardous beyond certain thresholds. The project is foreseen to strengthen the knowledge exchange and cooperation between the Nordic countries and in the following phases address the urgent matter in EU. The goal of the project is to reducing the number of inhabitants in the Nordic countries that are exposed to odour and airborne allergens as well as other emissions from animal farming with possible health impact and to investigate to what extent the various countries have developed national strategies in order to control and regulate odour annoyance and allergen of dispersion from livestock farming.

Keywords: odour management, bioaerosols, emission from livestock, allergens, annoyance, regulation, health and life quality

Introduction

Thousands of different odour and bioaerosols compounds from livestock farming have been identified (Attwood P. et al. 2004). Odour from livestock farming is usually a mixture of many compounds (Avery R. et al. 2004). Some of these may enhance the effect of other compounds whereas others may eliminate each other. A number of compounds that may be difficult to detect separately might in some cases together give a strong smell. The human nose is able to distinguish about 10,000 different odour compounds. More than 200 odour compounds have been identified in manure. Although odour is in gas phase, some compounds may be associated with dust and can later evaporate.

All types of animal house hold may lead to odour problems, but pig production appears to be the most important cause of odour leading to annoyance problems (Eder W. et. al. 2006) – at least quantitavely. Another important concern is the potential health problems related to allergens and other harmful compounds emitted from farm animals. In this context especially allergens from horses have been in focus in a strong debate that has taken place in Sweden.

The origins of the bioaerosols are the animals themselves: their feed, stools and urine with some allergens from skin and hair. Additional components stem from insects and microorganisms thriving on the organic material in animal buildings. Disinfectants and other agents applied to the environment are also present, and may add to the adverse health effects of workers (Preller L. et al. 1995). Bacteria thrive in this environment and give origin to high concentrations of bacteria, endotoxins, and other bacterial components in the air. The fungal load in animal houses with concrete floors without litter is likely to originate primarily from outside air. For livestock raised on litter or animals fed on hay fungi probably originate to a great extent indoors. This is important, since fungal spores appear to be closer associated with the asthma prevalence in livestock farmers than endotoxins and more protective in individuals disposed for allergic diseases and more harmful in individuals not disposed for allergic diseases (Eduard W. et al. 2004).

Bioaerosols containing this type of components have repeatedly been found to induce lung function changes, upper airway and mucosal inflammation, symptoms and sys-

[1] National Environmental Research Institute (NERI) Department for Atmospheric Environment (ATMI) Aarhus, Denmark

[2] Aarhus University, Department for Environment and Occupational Health, Aarhus, Denmark

[3] Finnish Meteorological Institute, Air Quality Research, Helsinki, Finland

temic inflammatory reactions in adults exposed to them.

Airborne concentrations of live bacteria are also very high (Duchaine C. et al. 2000, Donham K. J. et al. 1986, Attwood P. et al. 1987). Gram-positive bacteria dominate this population as they easily represent 90-95% of the total bacteria. Experimentally, endotoxins are capable of inducing many of the symptoms associated with livestock exposure, including fever reactions as seen in organic dust toxic syndrome and farmer's lung and worsening of asthma with cough and breathlessness. Thus, it is not surprising, that endotoxins have drawn so much attraction. Several epidemiologic investigations have found that respirable endotoxin in farming environments were closer associated with adverse effects on the airways and immune system than were airborne dust levels (Wing S. et al. 2002).

National status of odour

In Denmark an increasing pig production has increased the odour problems over the last decade. Table 1 shows the number of private residences in Denmark that are placed in the vicinity of livestock farms over a certain size. This selection is based on the number of Animal Units per livestock farm. One Animal Unit is defined as the animals leading to an emission of 100 kg N/year. This is equal to 0.85 milking cow in stable, annual production of 36 slaughter pigs (equal to 9 in stable), or 2900 annually produced slaughter chickens. Larger livestock farms typically cover an area with a diameter of 100 m (radius 50 m), and the distances given in Table 1 should therefore be reduced by approximately 50 m. Thus about 6700 residences are placed within 300 m from a livestock farm with more than 249 Animal Units. It should here be noted that the figures in the table refer to number of houses in Denmark with a potential odour problem related to livestock farming.

Table 1:
Number of private residences in the vicinity of livestock farms in Denmark based on registry data per 31/12/2002 (Source: Steen Gyldenkærne Policy Analysis, NERI 2005)

	Size and number of livestock farms	
	>125 DE	>249 DE
	6238	963
Radius (meters)	Number of private houses	
100	8708	1258
150	13939	1953
250	28190	3983
350	46548	6699
500	84312	12561
1000	298966	53630

Odour problems are mainly related to manure and the emissions may have three different sources: from stables, manure storages and from out bringing to the fields. The odour from storages may be reduced significantly e.g. by covering of manure storage tanks. Out bringing takes place over relatively short periods of time, whereas stables have to be ventilated continuously. Stables may therefore emit odour during the entire year and research in this field has to a large part been devoted to regulation of ventilation and control of air flows inside the stables.

In Denmark the agriculture is in general more intense than in the other Nordic countries. The public concern about especially about odour and ammonia has also been high for several years putting a pressure on the political system for regulation of this area through legislation. A Danish Guideline for handling odour from livestock farming has been in preparation for a longer period of time. Currently this Guideline is awaiting the restructuring of Danish counties and municipalities which took place by 1. January 2007.

In Sweden there has recently been an increasing concern not only to odour problems but also concerning the release of allergens from livestock farming and how these releases of allergens affect the health of the population in the nearby surroundings of the farms. In Germany the "NILS" study has shown, that there is a detrimental effect on the lung function of living in the vicinity of many animal farms. The researchers showed, that for people exposed to > 20EU LPS m^{-3} there was a tendency to asthmatic patterns in lung function measures. Allergens from horses may to a higher extend than allergens from other farm animals be spread further away from the farm houses.

Finland has no formalised guidelines for odour. Applied principles are formed with certain limit values or with set back distances. Odour management obligations for odour emitting plants are set in regional EPA environmental permits. Often the emission limits are set and followed up locally resulting in limited predictability for farmers. For life stock operation, set back distances are usually applied.

Livestock farming has caused odour complaints in Finland, the impact of these activities is usually limited to less than 0.5 km, although large pig farms can cause significant annoyance depending on the volume and animal unit.

Due to environmental measures, the odour load and annoyance has generally diminished from industries, agricultural odour being an exception. The reason for this is that the production units in Finnish livestock production are significantly increasing as well as in the other Nordic countries. Large animal houses are built closer to dwelling houses and as a consequence, odour annoyance becomes significant (Beaman A. L. 1988).

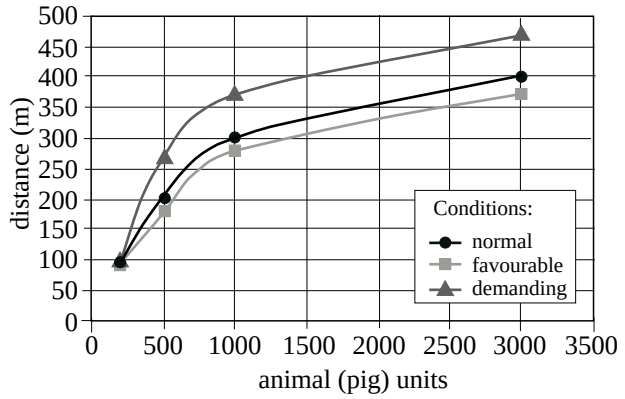
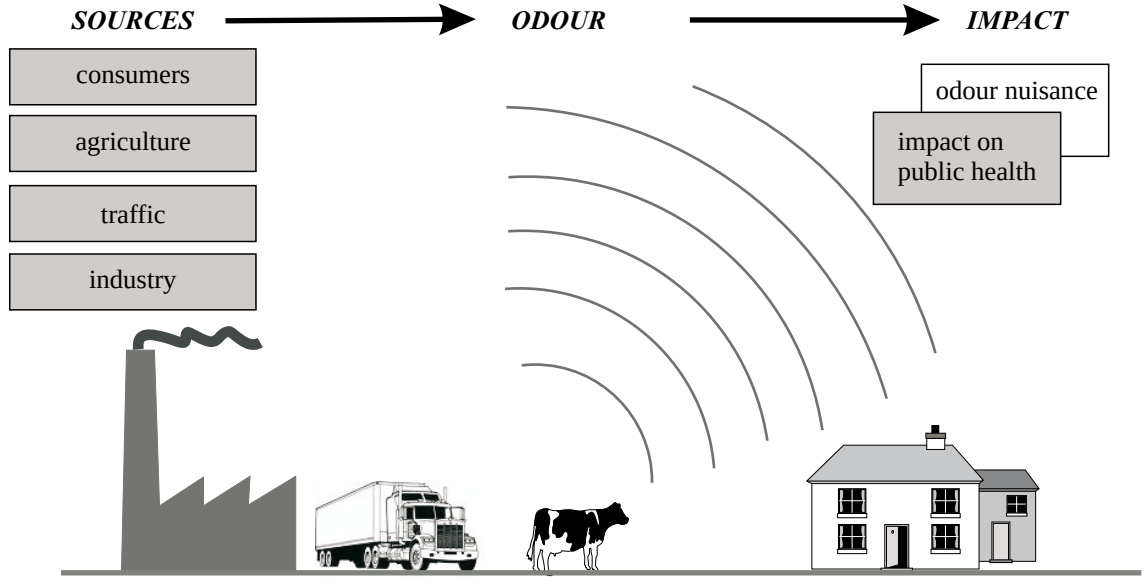


Figure 1:
Minimum distance between livestock units and sensible areas

In the nearest surrounding area of the production unit the odour occurrence levels are above 12 % of favourable condition calculated in yearly hours. The odour occurrence levels are decreasing as the distance from emission sources is increasing.



Source: RIVM

RIVM/ED/Oct02/0949

Figure 2:
Sources of odour

Distinction between odour nuisance and severe odour nuisance

The Dutch government uses two definitions for the environmental problem of odour nuisance: odour nuisance and severe odour nuisance. The concept of odour nuisance is based on the terminology used by Statistics Netherlands in its "Ongoing Survey of Living Conditions" (OSLC). The term 'severe odour nuisance' comes from the periodical nuisance survey conducted by the Dutch research institute TNO (also known as the 'questionnaire survey').

Odour nuisance (in the Statistics Netherlands definition) is defined as experiencing frequent or occasional nuisance from stench, in line with the questions asked in the OSLC. Sources of odour included in the survey are road traffic, industry or business, agriculture and open fires/multi-burners illustrated in figure 2.

Severe odour nuisance (in the definition given by TNO) is based on the question from the periodical nuisance survey of TNO about the extent to which people see a specific source in the living environment as a nuisance on a scale from of 1 (not a nuisance at all) to 10 (extreme nuisance). People giving answers in the 8 to 10 range are classified as experiencing 'severe nuisance' (Rantakrans E. et al. 1995).

It is not easy to compare the concepts because of the different ways the questions are formulated and the different definitions of the sources.

Allergens and health

It is a well known fact that exposure to the environment in swine confinement buildings is a cause of respiratory impairment and loss of lung function in farmers (Omland O. 2002, Omland O. et al. 2000, Preller L. et al. 1995b), Thorne, (Cormier Y. et al. 1991). Acute exposure to high amounts of dust from swine confinement buildings has

been shown to induce a neutrophilic pneumonitis.

Furthermore acute exposure of subjects has been shown to induce substantial more inflammation in subjects naïve to farming compared to farmers. Cattle and poultry are also known to cause both short and long term respiratory impairment among exposed workers. In addition there are reports on adverse effects on the respiratory system from exposures to other livestock such as sheep and horses. The common belief that odour is worse from swine than from cattle farms is supported by the greater emission rates from such buildings in Europe (Takai H. et al. 1998).

The airway diseases that can be caused by livestock exposures include development of allergic and non-allergic rhinitis, other upper airway and mucous membrane irritation symptoms, allergic and non-allergic asthma, aggravation of existing asthma, chronic obstructive pulmonary disease, hypersensitivity pneumonitis, and airway infections. Allergic alveolitis may be caused by exposure to mouldy hay and thus be related to although not directly caused by exposure to cattle and cows.

Odour alone has been shown to negatively affect immune function in neighbouring residential mediated via stress (Avery R. C. et al. 2004) but the isolated effect of odour has not been studied in livestock exposed workers. Allergens appear to play a limited role in industrialized farming environments such as in modern swine farming with low prevalences of allergic sensitization and allergic diseases. It cannot be ruled out that this is partly because of self selection out of the trade by individuals with atopic disposition. Most investigators agree that no single component or factor is responsible for the adverse health effects that occur after exposure to the animal farming environment. Rather the mixture of gases, dust particles, allergens, microbes and substances of microbial origin together induce the neutrophilic inflammation in the airways and the systemic changes in immune function.

Many different allergens of animal and plant origin are abundant in farming. In cattle breeders it has been shown that even several years after the last animal contact, there are significantly more allergens in the farmers houses, compared to other houses (Schulze A. 2006). For people having horses, it has also been observed, that their families are exposed to high amounts of horse allergen. This means that the allergens are stable over time, and can be transported from the stables to housing quarters of the farmers or horseback riders themselves. There is only very scarce information on the allergen concentrations in the area surrounding a horse stable or a cow-shed.

Exposure to high levels of endotoxin is particularly well documented in many types of farming, but other substances of microbiologic origin such as peptidoglycans and β-glucans are present in high concentrations. Airborne concentrations of live bacteria are also very high (Duchaine C. et al. 2000), Donham K. J. et al. 1986, Attwood P. et al. 1987a). Gram-positive bacteria dominate this population as they easily represent 90-95% of the total (dead as well as live) bacteria.

The origins of the bioaerosols are the animals themselves: their feed, stools and urine with some allergens from skin and hair. Additional components stem from insects and microorganisms thriving on the organic material in animal buildings. Disinfectants and other agents applied to the environment are also present, and may add to the adverse health effects of workers (Preller L. et al. 1995a). Bacteria thrive in this environment and give origin to high concentrations of bacteria, endotoxins, and other bacterial components in the air. The fungal load in animal houses with concrete floors without litter is likely to originate primarily from outside air (at least this is true for pigs on slatter). For livestock raised on litter (e.g. swine or cattle on chopped straw or on shavings) or animals fed on hay (such as horses) fungi probably originate to a great extent indoors. This is important, since fungal spores appear to be closer associated with the asthma prevalence in livestock farmers than endotoxins (more protective in atopics and more harmfull in non-atopics) (Eduard W. et al. 2004c). Gases evaporate from the manure pits underneath or in close adjunction to the swine buildings.

Bioaerosols containing this type of components have repeatedly been found to induce lung function changes, upper airway and mucosal inflammation, symptoms and systemic inflammatory reactions in adults exposed to them.

The effects on children's health are subject to some debate. On the one side there is evidence that the farming environment is protective against the development of allergies and some allergic disease and more so with animal exposure. On the other side, there is impelling evidence, that high concentrations of modern livestock operations in close vicinity of children's homes is associated with negative health effects and increased risk of lung disease including asthma-like symptoms. Children's exposure is likely to differ from that of adults with less exposure from inside concentrated animal buildings and more exposure to diesel exhaust and feed, grain and other dusts outside these buildings as well as odours. Livestock exposures even appear to be strongly protective against atopy in the prenatal period (Ege M. J. et al. 2006). Whether livestock exposures are protective or harmful depends on the genetic background of the exposed person and this is true both in childhood (Eder W. et al. 2006) and adulthood (Eduard W. et al. 2004a).

Differences in technology and climate is, however, likely to cause differences in qualities and quantities of exposures in residential areas. Importantly, it has been shown that whereas bioaerosol components such as gases and bacteria can be traced at long distances from CAFO's, they are di-

luted to minute amounts within very short distances of the ventilatory outlets. However, higher background levels of e.g. endotoxin can be found in rural areas with intensive livestock production than in urban areas (Schulze A. et al. 2006). With the current knowledge of mechanisms there is no reason to think that such low levels could have adverse health effects other than those caused by odour.

Quality of life, as indicated by the number of times residents could not open their windows or go outside even in nice weather, was found to be similar in residents in the vicinity of a cattle operation or far away from livestock but greatly reduced among residents near a hog operation (Wing S. et al. 2000). More wheezing has been observed among pupils at schools in the vicinity of confined swine feeding operations (Mirabelli M. C. et al. 2006).

Regulation and abatement

Since the end of the 1990's there was a strong need felt by the environmental authorities to improve the scientific basis of the odour regulations especially in the agricultural sector to increase their acceptance and effectiveness. (Cormier Y. 2004). This was further elaborated in a couple of larger research projects (Wing S. et al. 2002).

The concern of the public is growing in the Nordic countries. It would be beneficial in the Nordic countries to exchange knowledge about strategies behind environmental management and regulation of odour and allergens from livestock farming. A common Nordic strategy could be use a basis for recommendations to a new EU legislation in this field.

Emission factor data for pig and broiler production has been established for odour impact assessments. The emission data for pig production were slightly higher than results obtained in other countries, although in the same order of magnitude. Emission rates are influenced by a range of local factors including feed, manure management, building design and ventilation rate. Certain lack of unanimity in published odour emission data speaks up for the importance of using data obtained in representative national conditions. Finnish agricultural circumstances differ noticeable from those in Central Europe or North America concerning both climate and production methods.

Odour annoyance study has showed that people's reactions to pig and poultry odour are very different. Thus no clear indication for need for set back distances for mid-size the poultry production plants could be identified in the investigated plants. On the other hand, there seems to be a need for significant set back distances for large swine production units if no odour reducing measures implemented.

Dust emitted by housing units contributes to odour transport and plumes may have potential for transmitting diseases to other housing units or neighbouring people. Odour is combined with higher concentrations of endotoxins in the surroundings of a farm. This subject is currently being investigated with regard to potential effects on health of farmer families and neighbouring residential. At the same time, a Finnish study has indicated that newborns' exposure to microbes related to livestock farming diminishes the risk for the child to develop allergies (Omland O. et al. 2002).

Summary

Odour is one of the most remarkable environmental hazards caused by livestock farming. Odour is annoying people living in the neighborhood of farming units and odour inconveniencies may cause complaining in the vicinity of production units.

A major part of the on-going projects on air emissions from agricultural sources relates to monitoring and diminishing greenhouse gases. There is, however a need to revise the current general set of guidelines for livestock production and base them on the actual odour impact. Very little data is available e.g. on odour emissions from cow sheds and fur production.

A major odour source is the application of slurry in the fields. These intermittent fugitive odour sources are difficult to regulate and control. Investigation in the odour emission and annoyance arising from spreading slurry in the field would function as a base for further guidelines.

Exposure to high levels of endotoxin is particularly well documented in many types of farming, but other substances of microbiologic origin such as peptidoglycans and β-glucans are present in high concentrations.

Reduction of life quality in cities downstream from livestock farming some studies have been conducted to investigate possible negative effects of exposure from these facilities.

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Application of tensid mixed fog for seperation of organic/biologic aerosols

W. Haunold[1]

Abstract

The purification process in the atmosphere by fog and clouds is the most effective way to remove aerosol particles even in industrial applications. We have developed a seperation device which imitated the natural process by artificial fog produced for industrial use. Tensids added to the water are coating the tiny fog-droplets. This coating enable the fog to absorb organic emissions as well as biologic active aerosols high efficiently. Within a very short time of contact the adsorbed material will be removed together with the fog-droplets. This presentation will show first application at an industrial site.

Keywords : adsorption aerosol, waste-air purification, germ reduction, germ seperation

Introduction

New emission rules for the release of organic and biological active aerosols lead many companies in the industrial and agricultural sector to serious problems. CFU, (colony forming units) by bacteria, fungus and virus are hard to be removed out of the airstream. Our technique will help these copnpanies to catch the limits.

By using tiny, floatable fog-droplets a very efficient adsorption medium at low costs is created.

Droplets produced with with special laser drilled nozzles at 50 – 70 bar pressure.

They have a diameter of 5 – 20 micron, like natural fog.

The fog is airborne and therefore remains in the air-mass up to several minutes (figure 1).

1 gram of water dispersed as fog has a surface area of approximately 0.5 m². Aqueous fog can be acidic and alkaline activated and is in this form very efficient to reduce gases like H_2S and NH_3 in the exhaust air. The time needed to neutralize pH active gases with pH activated fog and is less than 1 second.

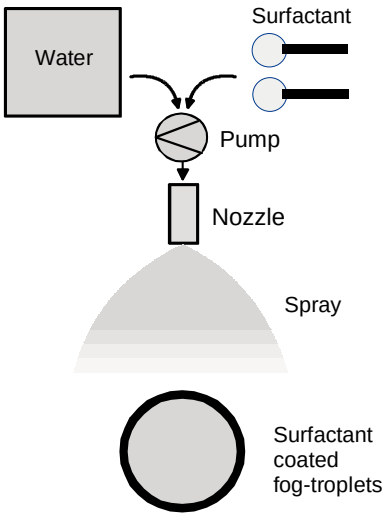


Figure 1:
Water containing an mixture of surfactants is sprayed by high pressure nozzles to generate a fog

The efficiency of adsoption of organic and odorus substances are strongly increased by mixing of only one percent of the patented additive tensids in fog droplets.

[1] Institute of Atmosphere and Environment, WG Prof. Jaeschke, University Frankfurt, Germany

Tensides are amphiphile molecules which combine a lipophilic and a hydrophilic part (figure 2).

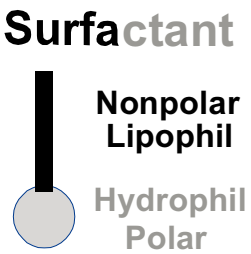


Figure 2:
Structure of a tensids

Tensids settle rather fast on the fog-droplets surface and cause an organic, lipophilic skin around the droplet.

The composition of tensids can be optimized for different types of emission „Tailer made Fog“. Even high hydrophobic substances are adsorbed.

The adsorption efficiency relays on the total surface area of the fog. The surface area is controlled by the droplet size and the total mass of fog water

Aerosol particles and organic molecules are reaching the surface of fog droplets by turbulence and molecular diffusion motion, where they were adsorbed (figure 3).

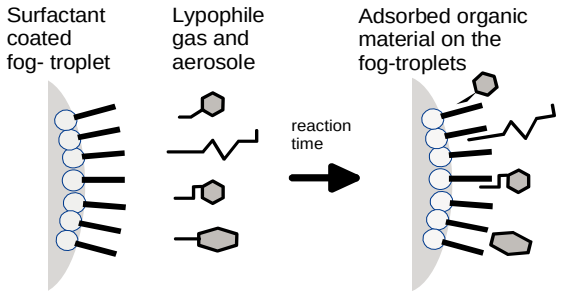


Figure 3:
Organic molecules and aerosol particles are connecting on the surface of the droplets where they are absorbed

Figure 4 shows the efficiency of the procedure as function of the droplet size and time of contact. So far tests were performed with fog droplets in the size range of 1 up to 1000 µm.

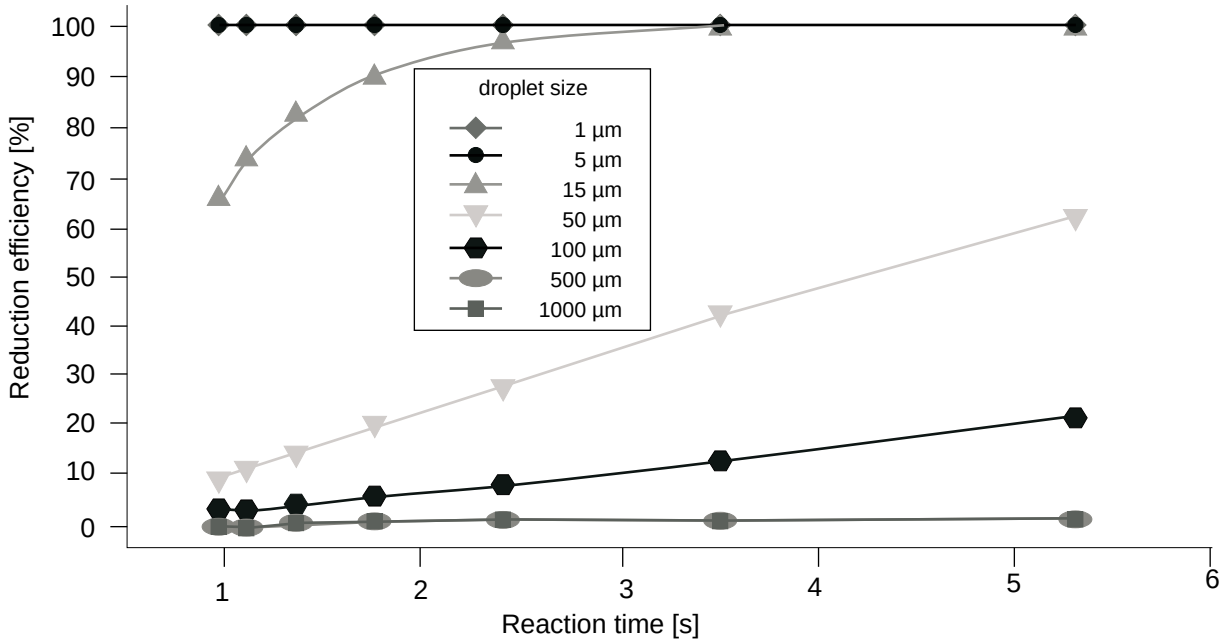


Figure 4:
Efficeiency of smell reduction as function of time of contact and droplet size

Pollutants absorbed by fog-droplets can be scavenged by the aid of demisters.

With this set up, air-pollutants are separated from the air together with the reagents (figure 5).

A sterilisation of the germs is not necessary they are washed out by the demistor.

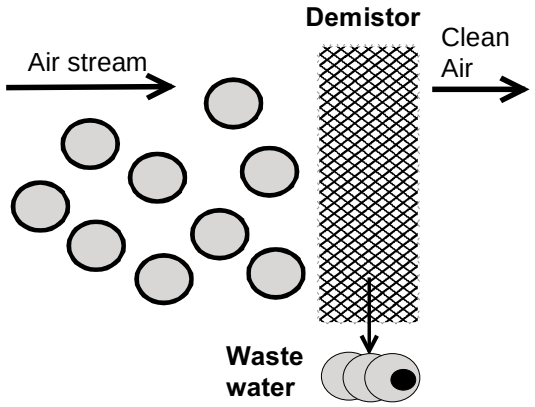


Figure 5:
Separation of fog and germs

10 g of fog activated with tensids per m³ waste air is able to eliminate more than 80 % of colony forming micro organisms within 5 seconds of contact. After contact the fog is separated with a demistor and all absorbed aerosols are removed. The result is a germ loaded liquid to be drained off in the reaction container and is run off into the sewagesystem (figure 6).

Sofar installed systems were build in customary containers. The fog is created by applying fog-nozzles at the air-inlet of the container. The containers are finished with stainless steel sheet-metal and can handle up to 50.000 m³ air flow per hour for germ reduction. Smaller units for tests are available (figure 7 and 8).

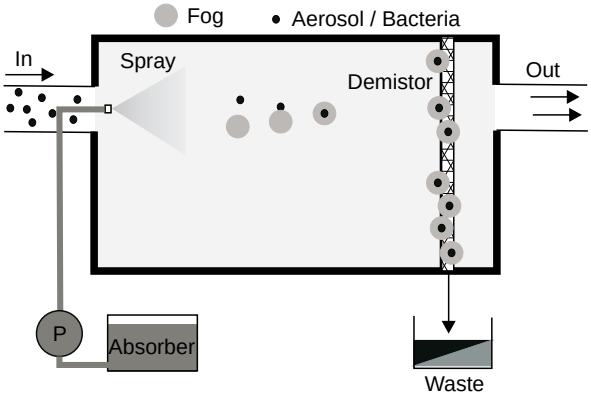
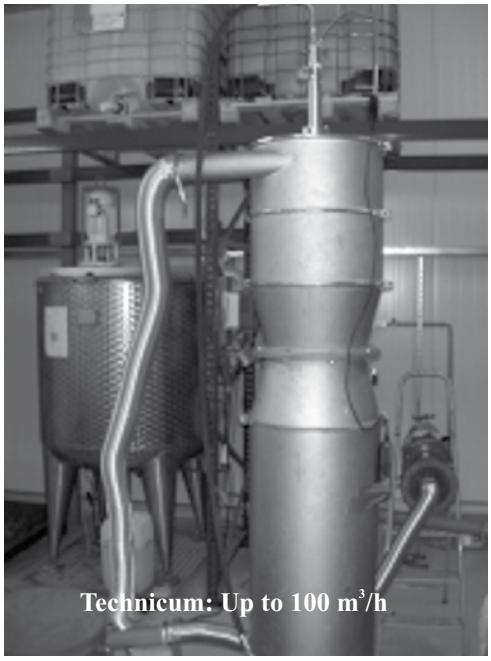


Figure 6:
Schematic diagram of the adsorption-container

Figure 7:
Setup with standard container (20 Feet), air flow 30 000 m³/h, water 300 l/h and 40 Nozzle; picture: Fog Systems

Table 1:
CFU number concentration taken before and after the separation container.
Values certificated by SGS Institute Fresenius

Before container		After container	
Cladosporium spp.	7500	Cladosporium	1375
Penicillium spp.	2500	Sterile colonies	63
Aspergillus flavus	1750	Acremonium sp.	25
Aspergillus fumigatus	1625	Alternaria sp.	25
Aspergillus niger	1250	Botrytis sp.	25
Aspergillus nidulans	375	Aspergillus fumigatus	<25
Sterile colonies	375	Aureobasidium pullulans	<25
Sum	**15375**	Penicilium sp.	<25
		Sum	**1558**



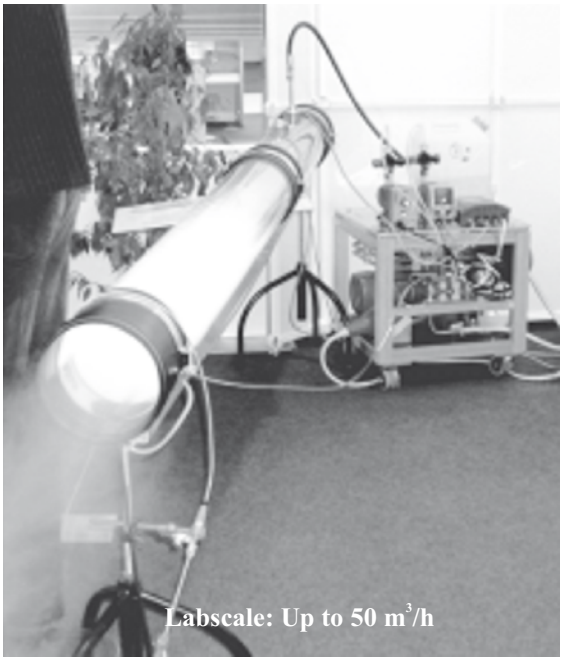


Figure 8:
Tests and measurements are possible in various fields of application and in different scales

The effectiveness of the procedure was shown predominantly so far in the field of smell reduction.

First results in the industrial and laboratory range show very high efficiencies of germ seperation.

Coming research projects shall show the efficiency of the system with virus infected waste air-treatment.

Table 2:
References and Projects

Life stock	Test in pig farms (>1000 animals) *pH <7 / 2% surfactants reduction 95% GE*
Textile processing	Burned cotton *pH mix / 2% surfactants reduction >94% GE*
Emissions from mixing vessels	Bitumen + Oil + mineral dust *pH >7 / 2 % surfactants reduction 96% GE*
Emissions from steel plants	Gaseous comp. from selective process steps *pH >7 / 2% surfactants reduction 96% GE*
Waste treatment site	Decomposition of gases (aerob and anaerob) *pH mix / 2% surfactants reduction 85% GE*
Waste treatment site	Germs in sorting area for waste goods *pH < 7 / 2% surfactants reduction KBE >90%*

GE = Odor Units

KBE = CFU

Acknowledgement

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Eur. Pat. 0 972 556 A1 „Adsorption von hydrophoben Gaskomponenten und/oder Aerosolen aus einer Gasphase“ CLIMAROTEC GmbH Bad Homburg v.d.H. (Patent erteilt 22.12.2004)

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Airborne dust control for floor housing systems for laying hens

G. Gustafsson[1] and E. von Wachenfelt[1]

Abstract

Floor housing systems for laying hens are being re-established in Sweden. Compared to traditional cage systems, the air in floor housing systems may be more polluted with dust.

Investigations about how different factors affect concentration and generation of dust in a floor housing system for laying hens have therefore been carried out at JBT:s research station Alnarp Södergård. A climate chamber was equipped with a floor housing system. How age of hens, storage time of manure, ventilation rate and bedding materials affected concentration and release of dust were investigated during two production cycles.

The concentration and generation of dust as well as the efficiency of different dust reducing measures were investigated and analysed in order to improve the understanding of how different factors influence dust conditions in these housing systems.

Settling of dust was a more important mechanism in the mass balance of dust than ventilation rate which reduced the influence of ventilation rate as a dust reducing measure. A major part of the generated dust settled on different surfaces inside the building. The settling rate of dust was affected by the concentration of dust in the air. The settled amount of dust also stood in relation to the floor area of the stable. An increased ventilation rate had a limited effect on the concentration of total dust due to the importance of the settling of the dust.

Dust release was also investigated when using six different bedding materials, namely; gravel, clay pellets, peat, wood shavings, chopped straw and chopped paper. Clay pellets and peat resulted in lowest concentrations of dust.

Automatic spraying of small droplets of water reduced the dust concentration in four trials with different bedding materials (chopped paper, clay pellets, peat and wood shavings).

Spraying a mixture of rape seed oil in water was also effective with an automatic spraying system.

From our findings from the investigation we will recommend using clay pellets as bedding material and using a sprinkler system spraying water droplets frequently in floor housing systems for laying hens.

Keywords: laying hens, dust, climate, ventilation, bedding

1 Introduction

Hartung J. (1998) states that poultry house air has much higher dust concentrations than for other animals. Seedorf J. (2000) demonstrated that particle emission in fowl keeping was 22 times higher (3165 mg h^{-1} per livestock unit) than for cattle keeping and four times higher than for pig keeping.

Regarding laying hens, floor housing systems are being re-established in Sweden since animal welfare legislation stipulates that systems for laying hens must include laying nests and perches and provide access to litter. Compared to traditional cage systems, the air in floor housing systems may be more polluted with dust because of high activity and more bedding material (Gustafsson G. et al. 1990, Hauser R. H. 1990, Lyngtveit T. 1992, Drost H. et al. 1992, 1993, Groot Koerkamp P. et al. 1993). The hygienic threshold limit value for dust for occupational safety and health (Swedish National Board of Occupational Safety and Health, 2000) of 5 mg m^{-3} is often exceeded during work operations in floor housing systems for laying hens. Whyte R. T. et al. (1993) reported that the average inspirable fraction breathed by poultry stockmen ranged from 2.1 to 28.5 mg m^{-3} for a complete working day.

In investigations about concentrations and emissions of airborne dust in Northern Europe (Takai H. et al. 1998) was it concluded that both inhalable and respirable dust concentrations were higher in percheries than in houses for caged layers. Ellen H. H. et al. (2000) reported that dust concentrations in perchery and aviary housing systems often were four to five times higher than in cage systems. Factors affecting dust concentrations are animal category, activity, bedding materials and season.

Whyte R. T. (2002) reported that average inhalable dust exposure by poultry stockmen for a complete working day in free range systems was 10.8 mg m^{-3} compared to 4.8 mg m^{-3} in cage systems.

Guarino M. et al. (1999) found that dust concentration in an enclosed laying house was significantly higher during periods with feed distribution and scraper cleaning than during the night.

Donham K. et al. (1999) reported a threshold concentration of 2.4 mg m^{-3} for human health inside poultry buildings.

In order to study the generation and concentrations of dust

[1] Swedish University of Agricultural Sciences, Department of Agricultural Biosystems and Technology, Sweden

and their correlation to other factors a small scale poultry house (climate chamber) was equipped with a floor housing system. Between 333 and 392 hens were kept in the system during the investigations.

The objective of the investigations was to evaluate the influence of following factors on dust concentration and generation:

* Age of hens
* Storage of manure
* Ventilation rate
* Bedding materials
* Fogging water droplets
* Spraying a rape seed oil mixture

2 Mass balance of dust

The mean generation of dust can be described as:

$$p = q (C_o - C_i) + S A \quad (1)$$

where: p is the production of dust in mg h^{-1} ;
C_o and C_i are the total dust concentrations in air outlets and inlets in mg m^{-3} ;
S is the settling rate of dust on floor surfaces in mg $m^{-2}h^{-1}$; and A is the area of the floor in m^2.

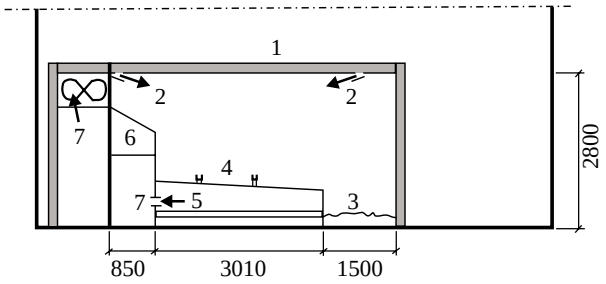
3 Materials and methods

How age of hens, storage time of manure, ventilation rate, bedding materials and spraying water droplets or an rape seed oil mixture affected generation and concentration of dust were investigated during two production cycles at JBT:s research station Alnarp Södergård.

3.1 Housing system

The investigations were carried out in a climate chamber equipped with a floor housing system, figure 1. The chamber was surrounded by a temperature controlled air space where the air temperature and supply air temperature to the chamber were varied between 0 and 16 º C.

The housing system contained a bedding area, a manure bin area with manure conveyors below a draining floor and laying nests which were placed close to one of the walls. The bedding area was 1.5 m wide where six different bedding materials were investigated successively, namely; gravel, clay pellets, peat, wood shavings, chopped straw and chopped paper. The rest part of the floor which was 3.0 m wide was elevated to a height of 0.6 m and equipped with a draining floor.



1. Climate controlled air space
2. Air inlet
3 Bedding
4. Draining floor
5. Conveyors
6. Nests
7. Air outlet

Figure 1:
The climate chamber equipped with a floor housing system

3.2 Ventilation and climate control

Mechanical ventilation was provided by a negative pressure system. Air inlets in the ceiling provided the chamber with supply air from the climate controlled area surrounding the chamber. In these studies an exhaust fan removed air at floor level close to the manure conveyors via a slit in a duct below the laying nests.

The temperature inside the chamber was kept at a constant level of 20 to 21 °C during each trial. The ventilation rate could thereby also be kept constant. The constant air temperature was maintained by controlling the amount of extra heat from heat pipes in the chamber.

The ventilation rate was manually regulated with a damper in an exhaust air duct.

3.3 Measurements

The ventilation rate was calculated from air velocities measured two times per trial in 5 positions of the cross section of the exhaust air duct (ϕ 400 mm) by using a hot wire anemometer (GGA- 65P, Alnor Instrument CO, Skokie, Illinois, USA).

The efficiencies of different treatments were determined by gravimetric measurements of sampled dust masses on 37 mm diameter dust filters (Millipore with an air flow rate of 1.9 l/min) in SKC cassettes located in the middle of the barn at 1.7 m height above the floor (breathing zone of humans) but also in the exhaust air. Each sampling period was 3 - 4 days. Sampling was done 15 minutes each hour controlled by a timer. Settled dust, which was sampled on five 0.230 m^2 settling plates located at a height of 2.0 m (height was chosen so that they could not be moved during work operations) was also gravimetrically determined. Each measurement was carried out over a period of

3 - 4 days in order to collect enough dust on the settling plates. Different treatments were compared to reference values measured before and after the treatments.

The generation of dust was determined according to equation 1.

3.4 Analyses

Different measures to reduce the generation and concentration of dust was analysed by using the following properties in the mass balance in equation 1: averages of total dust concentrations C_o measured in the exhaust air; average of settling rate of dust on settling plates S; generation of dust p as defined by equation 1;. Concentration in inlet air, C_i, was assumed neglectable compared to C_o.

3.5 Investigations

The influence of age of the hens on release of dust was investigated during two production cycles.

The influence of increasing storage time of manure in the bedding area was investigated during periods with daily manure removal from the conveyors.

The influence of ventilation rate was determined at constant air temperature (20 to 21 ºC) at varying ventilation rates.

Six different bedding materials were investigated namely; gravel, clay pellets, peat, wood shavings, chopped straw and chopped paper at ventilation rates in the range of 1.04 – 1.13 m^3 hen^{-1} h^{-1}.

How different amounts of water which was fogged influenced dust concentration was investigated using four different bedding materials (chopped paper, clay pellets, peat and wood shavings). Full cone nozzles (Fulljet 5LVS) were used. The equipment for controlling the spraying time is presented in figure 2.

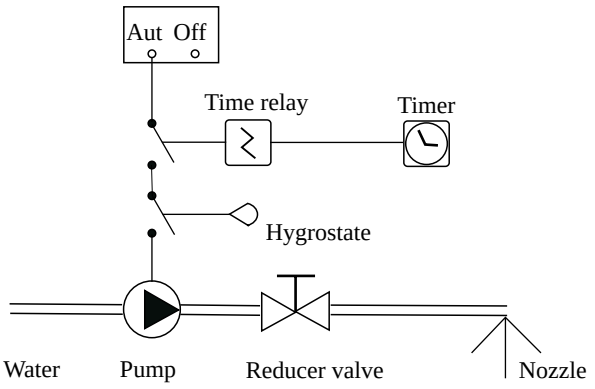


Figure 2.
Equipment for control of spraying time and intervals for the spraying nozzle

Spraying was done twice per hour during the light period 4.30 a.m. – 5.30 p.m.. Different amounts of water were investigated by varying the spraying time with a time relay.

The effect on dust concentration by fogging water droplets was compared with reference periods without any fogging.

The effect of spraying different amounts of a mixture of 10% rapeseed oil in water on dust concentration was also investigated. The mixture was supplied once per day with the same equipment as for water droplets.

4 Results and discussion

4.1 Age of hens

It could not be proved that the age of the hens had any influence on the generation of dust.

Obviously the dependence of age in this type of housing system for layers differs with the generation of broilers were it has been reported that the generation of dust increases with age and weight of chickens (Gustafsson G. et al. 1990). An explanation may be that the weight of the hens is relatively constant during the production period.

4.2 Storage of manure

The storage time of manure in the bedding had no significant influence on the generation of dust. Accumulation of manure in the bedding seems therefore not to be the major source for the generation of dust.

4.3 Ventilation rate

The ventilation rate had a limited diluting effect on dust concentration, figure 3.

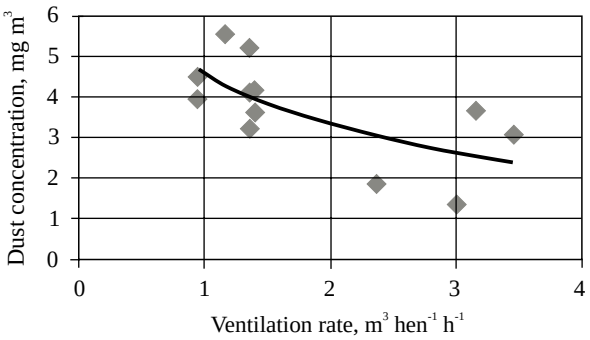


Figure 3:
Total dust concentrations at different ventilation rates in a trial with gravel as bedding material

Even if dust concentration decreased at increasing ventilation rate, it was not an ideal dilution depending on ventilation rate. The variations in dust concentrations were large which indicates that there were other factors as activ-

ity in the building environment which were more important for the dust concentration than ventilation rate. The ventilation rate did not either have any significant influence on the generation of dust. The average dust generation was 27.9 mg hen^{-1} h^{-1} when the ventilation rate was in the range of 0.9 – 3.4 m^3 hen^{-1} h^{-1}. The investigations showed that it was a limited amount of the dust generated which was exhausted with the ventilation air, figure 4. The reason was that the major part of the dust produced settled on different surfaces. This result is in accordance with results earlier reported for swine and chickens (Nilsson C. et al. 1987, Gustafsson G. et al. 1990).

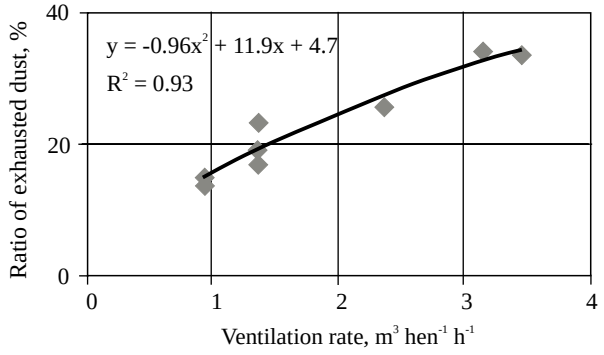


Figure 4
Ratio of exhausted dust at different ventilation rates in a trial with gravel as bedding material

4.4 Settling of dust

The settling rate of dust, S, was also analysed as function of the dust concentration in the air, see figure 5. The settling rate increased with increasing concentration.

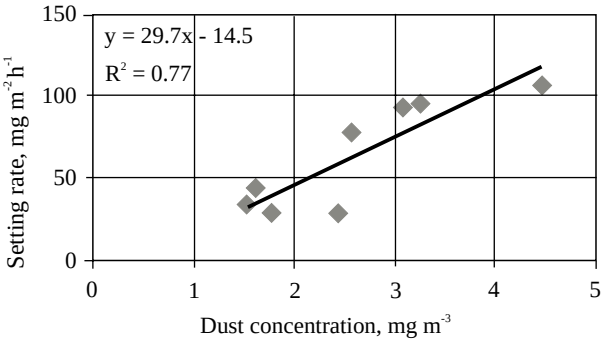


Figure 5:
Settling rate of dust as function of dust concentration when gravel was bedding material

4.5 Bedding materials

Total dust concentrations with different bedding materials are presented in table 1 when the ventilation rates were in the range of 1.04 – 1.13 m^3 hen^{-1} h^{-1}.

The level of concentration of dust was about the same for the bedding materials wood shavings, clay pellets, peat and chopped straw. Especially gravel resulted in higher concentrations, however, not statistically different.

Regarding the generation of dust was the picture about the same as for concentration of dust except for peat and chopped paper as bedding materials, table 2. The high dust production with peat depended on a high settling rate.

Table 1:
Total dust concentration (mg m^{-3}) with different bedding materials. Ventilation rate in the range of 1.04 – 1.13 m^3 hen^{-1} h^{-1}

Bedding:	Average mg m^{-3}	Minimum mg m^{-3}	Maximum mg m^{-3}
Gravel	4.7	4.0	5.5
Wood shavings	2.3	2.1	2.4
Clay pellets	1.8	1.7	1.9
Peat	1.7	1.2	2.3
Chopped straw	2.1	1.8	2.3
Chopped paper	2.6	2.2	2.9

Table 2:
Total dust production (mg hen^{-1} h^{-1}) with different bedding materials. Ventilation rate in the range of 1.04 – 1.13 m^3 hen^{-1} h^{-1}

Bedding:	Average mg hen^{-1} h^{-1}	Minimum mg hen^{-1} h^{-1}	Maximum mg hen^{-1} h^{-1}
Gravel	27.4	26.6	28.3
Wood shavings	11.4	10.4	12.4
Clay pellets	8.5	7.6	9.8
Peat	21.7	12.1	35.3
Chopped straw	16.8	15.7	17.8
Chopped paper	22.7	18.5	25.5

4.6 Fogging water droplets

How different amounts of water which was fogged influenced dust concentration was investigated using four different bedding materials. The effect of fogging was analysed as relative values compared to the levels without any fogging. Fogging resulted in a considerable reduction of dust concentration in all trials. The reduction in dust concentration was improved when the amount of water increased, which is exemplified in figure 6.

Fogging water droplets is obviously an effective way of reducing dust concentration in this type of housing system. Von Wachenfelt E. (1999) has earlier reported up to 65% reduction by fogging water droplets in an aviary system for laying hens.

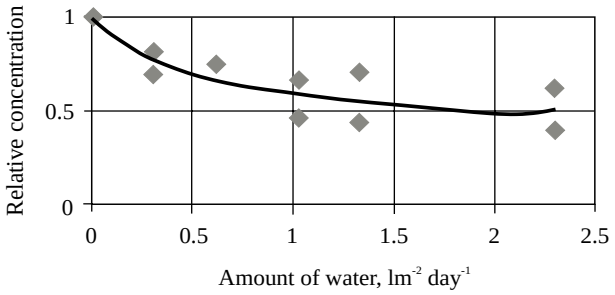


Figure 6:
Relative dust concentration (1.0 is reference level) at different amounts of water sprayed. Wood shavings was bedding material

4.7 Spraying a rape seed oil mixture

The effect of showering a mixture of 10 % rape seed oil in water on dust concentration was also investigated, figure 7. The mixture was showered with full cone nozzles located above the draining floor. Showering the oil mixture reduced the dust concentration with 30 to 50 %. The oil mixture had effect at as low amounts as 0.003 l m^{-2} day^{-1}.

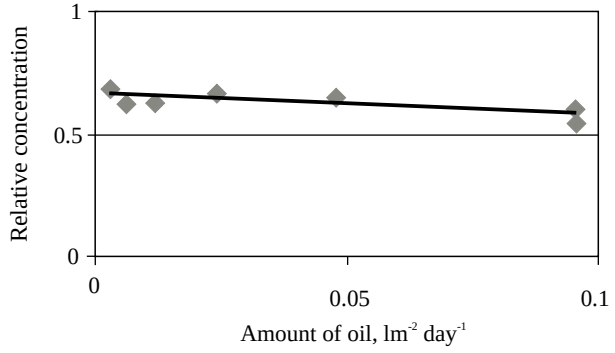


Figure 7:
Relative total dust concentration (1.0 is reference level) at different amounts of a sprayed oil mixture

5 Conclusions

The following conclusions can be drawn from the investigations:

- There were no ideal diluting effect of ventilation rate on dust concentration.
- A major part of the produced dust settled on different surfaces.
- Spraying water droplets or an oil mixture reduced dust concentration.
- The feather conditions were very good when water droplets or an oil mixture were sprayed.
- Bedding of clay pellets or peat generated the lowest concentrations.
- Gravel as bedding material generated the highest concentrations.

From our findings from the investigations we will recommend using clay pellets as bedding material and using a sprinkler system spraying water droplets frequently in floor housing systems for laying hens.

6 Acknowledgements

Financial support from the Swedish Farmers´ Foundation for Research and the Swedish Board of Agriculture is gratefully acknowledged.

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Measuring particle emissions in and from a polish cattle house

T. Hinz[1], S. Linke[1], P. Bittner[1], J. Karlowski[2], and T. Kolodziejczyk[2]

Abstract

After ammonia PM comes more and more in the focus of international strategies of air pollution control. Additional PM is a factor of air quality inside the buildings related to health and welfare of farmers and animals.

One of the main sources are livestock houses of poultry and minor pigs and cattle. Annual emissions will be calculated from activity data (animal places or numbers) and emission factors. These emission factors must be determined by measurements. In contrast to ammonia there are no models available to determine particle emissions.

Effects of particles on individuals and the dispersion of particles in the ambient air strongly depend on the size that means ultimately the mass of the single particles. Various fractions are defined by different definitions. The paper gives the most important definitions and procedures how to measure particles in a size selective way.

In a project of bilateral German-Polish cooperation in agricultural research, measurements in a dairy house were carried out in summer and winter 2006. Flow rates and PM concentration were measured inside the houses and in the exhaust of a force ventilated stable.

Concentration of TSP (total dust) was below 0.6 mg/m³ with higher values in summer than in winter, whereby measures inside the stable were higher than in the exhaust flow. Emissions consist of 100 % PM10.

One aim of the studies was to give an improvement of emission factors used in emission inventory, but this spot measurements will give only the impression that dependent on the management these factors may be lower than the usually used.

Keywords: dairy cattle, PM, emissions, emission factors, air quality

Introduction

There is an increasing need to control emissions of particulate matter (PM). Agriculture is a substantial source of PM emissions. PM also reduces the air quality within livestock buildings with implications for the health and welfare of farmers and animals. Effects of particles on individuals and their dispersion in the air depend on different parameters but strongly on their size and mass. Different target–oriented definitions are used (ISO 1996, US EPA2001), figure 1.

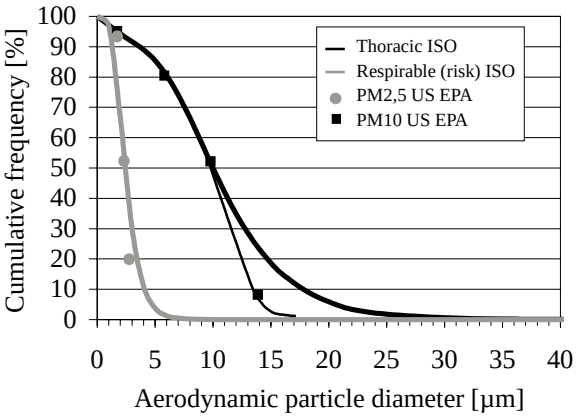

Figure 1:
Definitions of particle fractions

In a bilateral German- Polish research projects, measurements were carried out in force-ventilated building housing dairy cows. Concentration of total dust (TSP) and the PM10-fractions inside the house and the air flows through the exhaust ducts were measured and emission factors calculated. These values may be useful to estimate the size of emissions and the influencing parameters.

Materials and Methods

Investigations were done on a dairy farm in the Konin region of Poland. 64 cows with an average milk yield of 9200 kg milk per year and cow were kept in a building of 46.25m · 12 m equipped with four temperature controlled axial fans with a nominal maximum flow of 5950 m³/h each. Figure 2 shows the floor plan of the stable and the locations of the fans in the roof.

[1] Institute for Technology and Biosystems Engineering, Federal Agricultural Research Centre, Braunschweig, Germany

[2] Institute for Buildings Mechanisation and Electrification of Agriculture, Poznan, Poland

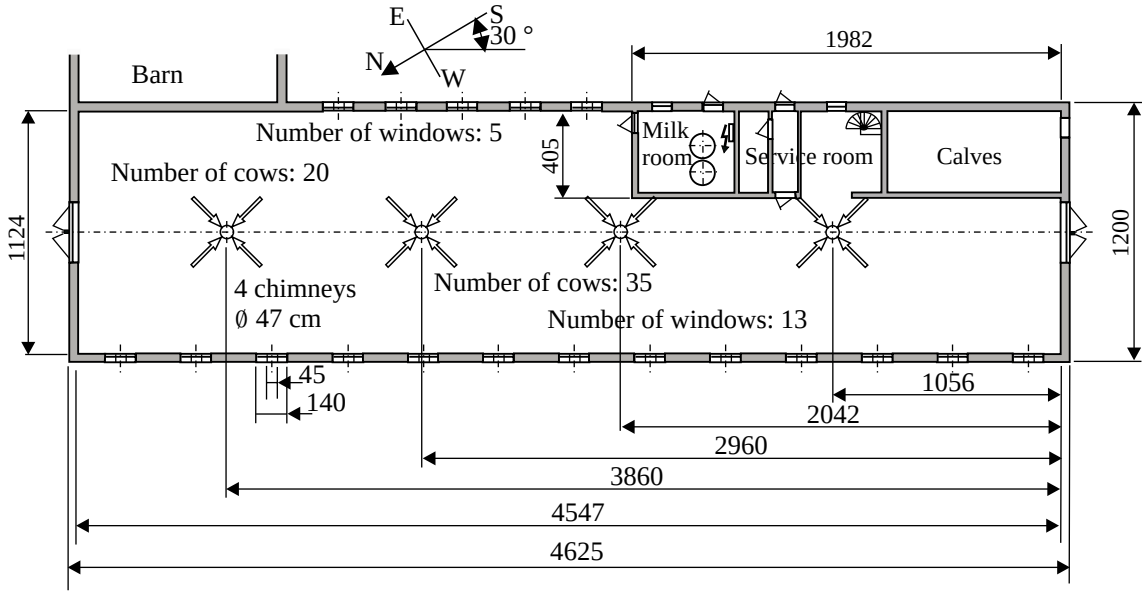

Figure 2:
Floor plan of the cattle house with exhaust openings location

There was a passage in the corridor used for forage distribution. On both sides of it were forage tables and two rows of chained cows' boxes with straw bedding. Straw manure is removed by mechanical grabbers of an electrically-powered machine once a day.

Fresh air entered the cow house through windows with variable openings. The degree of declination of the windows was adjusted manually upon evaluation of atmospheric conditions as given in figure 3.

Measuring particle emissions from force ventilated stables requires isokinetic sampling in the duct to get the

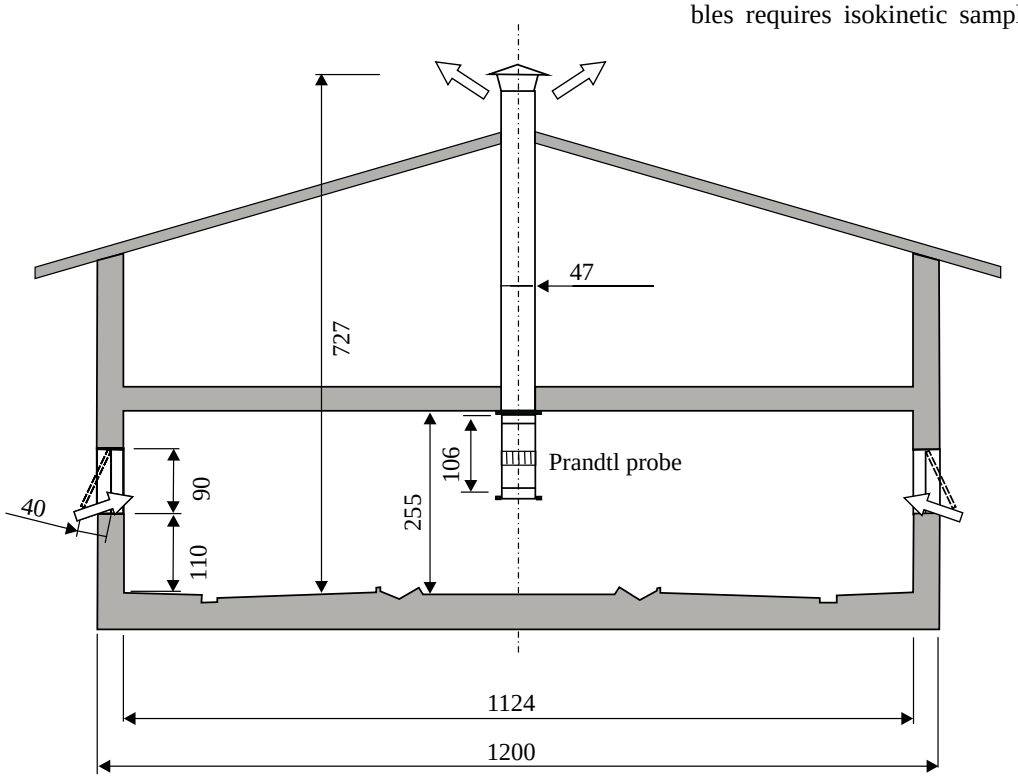

Figure 3:
Cross section of the cattle house

representative information about particle mass concentration in the balance area; figure 4 (Hinz T. 2005).

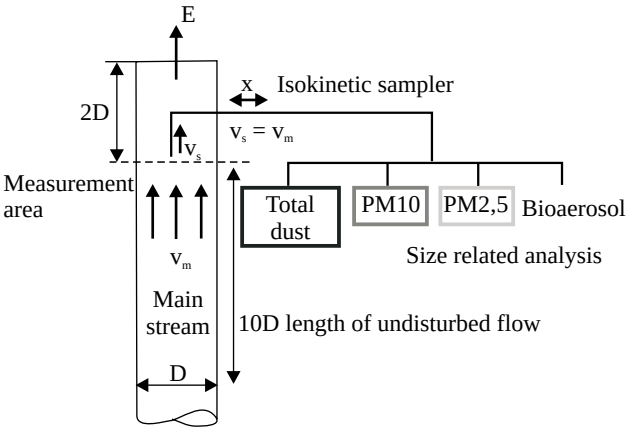

Figure 4:
Scheme of isokinetic sampling

To ensure the isokinetic condition at each measuring location, flow velocity was monitored using a hot wire anemometer.

To asses the emissions actual flow rates through all the ducts must be known. It was not possible to do so during the measuring campaign. Pre- investigations were carried out to get knowledge about flow distribution in the four ducts. For this purpose air velocity profiles were measured using a Prandtl-probe. The measuring positions inside a duct, which were also basically used to arrange the PM sampler, are given in figure 5.

At eight positions in two cross sections each grid measurements of air veloctiy were used to calculate average air flow rates and to calibrate voltage control of the ventilation system.

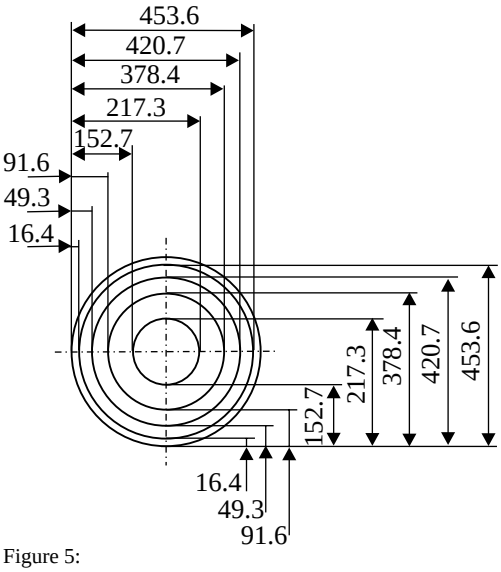

Figure 5:
Array of measuring locations to calculate exhaust air flow

Samples inside the buildings were collected with a sampling velocity of 1.25 m/s according to the conditions at working place.

In both cases inside the stable and in exhaust a conventional gravimetric filter procedure served as reference of total dust. To determine the complete size distribution a high-volume sampler was used with a pre-separator to separate large particles. The coarse fraction was analysed with light diffraction method. A scattering light monitor was installed in the flow behind the cyclone separator in order to determine the passing through of fine particles. For control and calibration purposes an absolute filter collected the fine particle fraction. Figure 6 shows the complete ensemble of inside measurements.

Figure 6:
Set of instruments to measure concentration and particle size inside the stable

Results and Discussion

Results will be presented for the pre- investigations to calibrate the ventilation system, the air flow rates and the measurements of concentration and particle size inside the stable in comparison with the emissions.

Monitoring of air flow rate in the ventilation system has been replaced by monitoring of the supplying voltage of the fans after previous assessment of air flow in each chimney separately. The sum of capacities of all four chimneys represents total running capacity of the ventilation system that is a function of supplying voltage delivered by the speed controller.

The coefficient of determination R^2 = 0.9906 is sufficient. The function was used in further calculations for estimating the air exchange in the stable.

From March 2006 to 2007 the ventilation rate changed between approximately 8000 m³/h and 12000 m³/h measured in the hot July 2006. The columns of figure 8 give the course of the year.

The investigations in dust concentration and dust emissions were carried in July and November 2006. Figure 9 shows the measured airflows for the respective days, which do not differ widely from the monthly averages.

In July the air exchange raised up to approximately 11000 m³/h while in November an average of 9000 m³/h was measured caused by the lower temperature.

Table 1.
Air flow through the chimneys as a function of supplying voltage

Fan	Flow rate	Supplying voltage [V]								
		70	90	110	130	150	170	190	210	230
K-1	[m³/h]	1 858	1 768	2 358	2 537	2 865	3 353	3 459	3 213	3 279
K-2	[m³/h]	1 440	1 745	2 256	2 565	2 787	3 096	3 291	3 412	3 478
K-3	[m³/h]	1 290	1 624	2 167	2 524	2 742	2 893	3 039	3 204	3 342
K-4	[m³/h]	1 628	1 823	2 260	2 518	2 756	2 967	3 099	3 205	3 302
Total	**[m³/h]**	**6 217**	**6 960**	**9 041**	**10 144**	**11 150**	**12 308**	**12 888**	**13 034**	**13 402**

Total performance of the ventilation system is shown in the diagram below, figure 7.

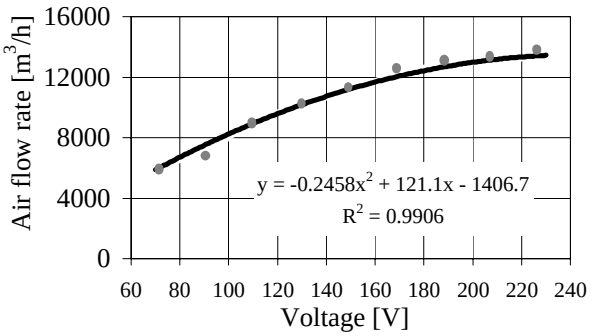

Figure 7:
Calibration of the ventilation control, total air flow rate of the 4 fans

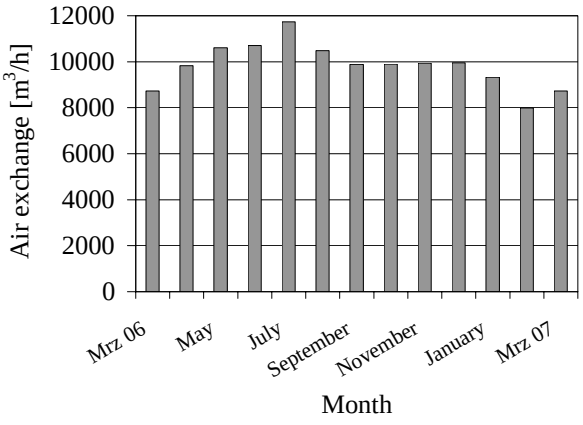

Figure 8:
Air exchange in the cow stable in the period from March 2006 until March 2007

Thus performance of the ventilation system is presented by the equation:

$$V = -0.246 \cdot U^2 + 121.1 \cdot U - 1406.7 \quad [m^3\ h^{-1}]$$

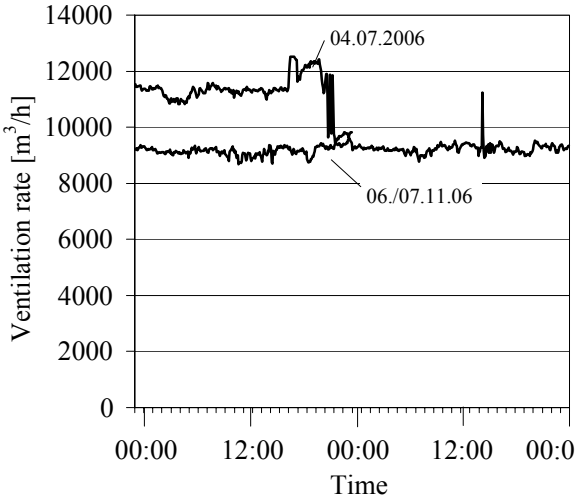

Figure 9:
Air exchange in the cow stable at 04.07.2006 and 06./07.11.2006

The concerned concentration of total dust (TSP) inside the stable, in the exhaust and the resulting emissions of TSP and PM10 are given in table 2.

Table 2:
Concentration and emissions in and from the stables

Type of animal / season / litter	C_{inside} [mg/m³]	$C_{exhaust}$ [mg/m³]	M_{TSP} [g/(animal/h)]	M_{PM10} [g/(animal/h)]
cow / summer / straw	0.550	0.188	0.033	n.a.
cow / winter / straw	0.198	0.064	0.009	0.008

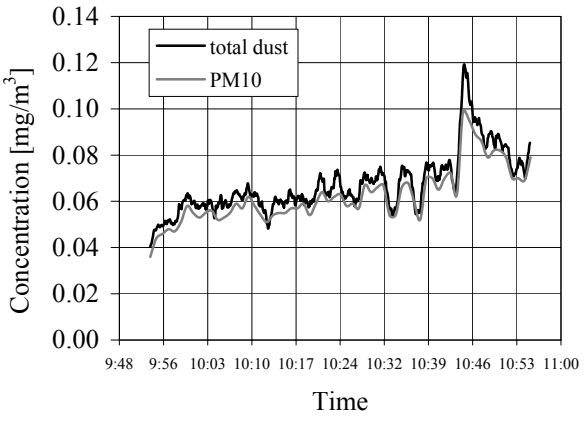

Figure 10:
TSP and PM10 concentration in the exhaust flow

In the cow stable the concentration was generally low with values <1 mg/m³. Depending on the climate conditions PM concentration and emissions were lower in November than in June. In both cases the concentration inside the stable at 1.5 m above ground was 3 times higher than the concentration in the exhaust flow.

Particle size distribution and with this the ratio of PM10 differ essentially between the air inside the stable and the exhaust flow, figure 10 and figure 11.

PM emissions consist of the fraction PM10 only. In contrast to this TSP concentration inside the building was much higher than for PM10, figure 11.

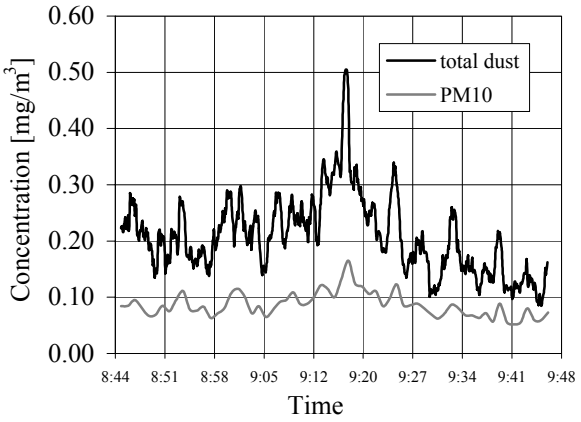

Figure 11:
TSP and PM10 concentration inside the stable

Depending on the principle the used light scattering monitor is not able to classify coarse particles >20 µm. By cyclone separation TSP was split to 5 % fine and 95 % coarse fraction. Particle size analyses of the coarse fraction show a wide distribution with particle sizes up to 300 µm and only a little proportion of approximately 15 % of PM10, figure 12. In total only 20 % of TSP form PM10 in the stable.

The differences between concentration and size distribution confirm the necessity to measure in the exhaust flow to get the right emissions.

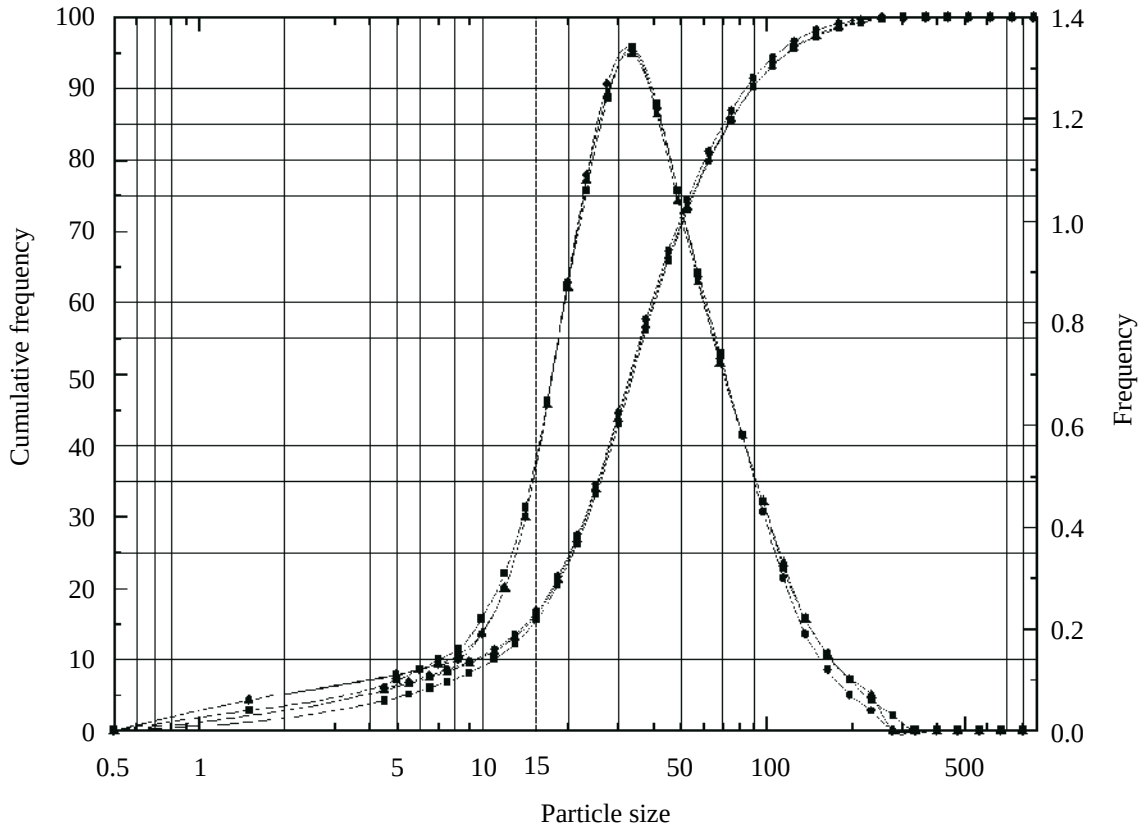

Figure 12:
Size distribution of the separated coarse fraction in the stable air

Conclusion

On a dairy cattle farm in Konin, Poland, PM emissions in and from a cattle house were investigated. Instrumentation was used to measure total dust and PM10 by gravimetric procedure and online monitoring.

An excellent management strategy by the farmer resulted in a very clean dairy house with very low concentration and emissions, which may be not the normal case.

In summer concentration was higher than in winter.

Concentration inside the building was higher than in the exhaust flow.

The ratio PM10/TSP was nearly 100 % in the emission flow, but 20 % only in the stable air.

References

Hinz T. (2005). Particulate matter emissions as a part of air pollution control in agriculture. Definitions, sources, measurements, 63-70. In (eds.) Kuczynski T., Dämmgen U., Webb J., Myczko A.(1996). Emissions from European agriculture.Wageningen Academic Publishers

ISO 7708 Publication date: 1996-01 Air quality- Particle size fraction definitions for health- related sampling.

US EPA: Code of Federal Regulations; PM10, 2001a PM2.5, 2001c

Measurement, analysis, and modeling of fine particulate matter in high ammonia region of Eastern North Carolina, U.S.A.

V. P. Aneja[1], S. Goetz[1], and Y. Zhang[1]

Abstract:

An analysis of fine particulate data in eastern North Carolina is conducted in order to investigate the impact of hog industry and its emissions of ammonia into the atmosphere. The fine particulate data are simulated using ISORROPIA, an equilibrium thermodynamic model that simulates the gas and aerosol equilibrium of inorganic atmospheric species. The observational data analyses show that the major constituents of fine particulate matter (PM2.5) are organic carbon, elemental carbon, sulfate, nitrate, and ammonium. The observed PM2.5 concentration is positively correlated with temperature but anti-correlated with wind speed. The correlation between PM2.5 and wind direction at some locations suggests an impact of ammonia emissions from hog facilities on PM2.5 formation. The modeled results are in good agreement with observations, with slightly better agreement at urban sites than at rural sites. The predicted total inorganic PM concentrations are within 5 % of the observed values under conditions with median initial total PM species concentrations, median relative humidity (RH), and median temperature. Ambient conditions with high PM precursor concentrations, low temperature, and high relative humidity appear to favor the formation of the secondary PM.

Keywords: PM2.5, ammonia; hog industry; ISORROPIA; inorganic aerosols

Introduction

Particulate matter has become a relatively recent concern in the overall air quality of our environment. In 1997, the Environmental Protection Agency modified the National Ambient Air Quality Standards for particulate matter by dividing the total suspended particulate standard into two separate modes of particulates, fine (PM2.5) and coarse (PM10-2.5) particles, with the standards for fine particulates being 65 μg m^{-3} daily and 15 μg m^{-3} annually. The US EPA has recently tightened the daily-average standard for PM2.5 to be 35 μg m^{-3} The fine mode of PM is known to contribute to human respiratory problems, dry and wet acidic deposition, reduced visibility, and radiative forcing (US EPA, Office of Air & Radiation, 2005). PM2.5 is composed of primary and secondary pollutants; primary PM2.5 species may include organic carbon, elemental carbon, soil dust, ash, and sulfate. Secondary PM2.5 may include sulfate, nitrate, ammonium, and organic carbon, which are formed through the oxidation of their gas-phase precursors such as sulfur dioxide, nitrogen dioxide, ammonia, and volatile organic compounds.

In particular areas of the United States, ammonia and ammonium have become a significant contributor to total PM2.5 concentration. Ammonia can react with acidic compounds to form various aerosols such as ammonium nitrate (NH_4NO_3), ammonium chloride (NH_4Cl), ammonium sulfate ($(NH_4)_2SO_4$), and ammonium bisulfate (NH_4HSO_4). Globally, it is estimated that a total of 49.3 Tg of NH_3 is emitted into the atmosphere with 56 % of this total being anthropogenic. The largest contributor to these ammonia emissions is domestic animal waste decomposition, which accounts for 22 Tg NH_3 (Warneck P. 1988, Schlessinger W. H. et al. 1992, Crutzen P. J. et al. 1990, Duce R. et al. 1991). In the state of North Carolina alone, the largest source of ammonia emission is domestic animal waste (Aneja V. P. et al. 2001).

In recent years, the hog industry of North Carolina has experienced rapid growth. Between 1986 and 2005, the hog population expanded from 2.4 million up to 9.7 million, which makes it rank the second in terms of pig production by state nationwide (NCDA & CS, 2005). The swine in North Carolina are estimated to emit 68,540 tons of ammonia per year, which makes swine the largest contributor among all domesticated animals in North Carolina

[1] Department of Marine, Earth, and Atmospheric Sciences, North Carolina State University ,Raleigh, NC 27695-8208, USA

(Aneja V. P. et al. 1998). These swine are concentrated in the coastal plain region of the state or the southeast corner covering Bladen, Duplin, Greene, Lenoir, Sampson, and Wayne counties (Walker J. T. 1998).

A number of aerosol modules have been developed to simulate fine particulate matter. A particular area of focus has been studying the inorganic aerosols of fine particulate matter, which make up 25-50 % of total fine particulate matter (Grey H. A. et al. 1986). Some examples of these aerosol modules are MARS-A, SEQULIB, SCAPE2, EQUISOLV II, and AIM2, which have been thoroughly reviewed for their similarities and differences (Zhang Y. et al. 2000). ISORROPIA is a thermodynamic equilibrium model used for predicting the partitioning of major inorganic species between the gas phase and aerosol phase. This model was selected due to its efficiency in computation and its overall satisfactory performance against more comprehensive aerosol thermodynamic models. With an input of temperature, relative humidity, and the total (gas + aerosol) concentrations of sodium, ammonium, nitrate, chloride, and sulfate, ISORROPIA predicts how much the total amount will be in the gas and aerosol phases (Nenes A. et al. 1998 1999).

Measurement and Modeling Methods

PM2.5 observational data was obtained from the North Carolina Division of Air Quality (http://daq.state.nc.us/). This data consists of average daily values for seven sites in Eastern North Carolina between 2001 and early 2004. The exact specifications of the particulate data are listed in Table 1. Fayetteville and Raleigh are the urban sites, which are situated to the west of the majority of the hog facilities. Goldsboro, Kenansville, and Kinston are the rural sites, with Kenansville being both the smallest city and the most enclosed by the hog facilities. Jacksonville and Wilmington are two coastal sites with the hog facilities to the north and west of their positions. Figure 1 shows the locations of the seven sites in North Carolina and their relative positions to hog facilities (Blunden J. 2003). For all these sites, when the average daily value consists of less than 90 % of the individual hours reporting, the average daily data point is considered inaccurate and is discarded. Meteorological data was obtained for each site from the North Carolina State Climate Office (http://www.nc-climate.ncsu.edu). While PM2.5 data is available for all seven sites, speciated PM2.5 data is only available for two urban sites, Fayetteville and Raleigh, and one rural site, Kinston.

Table 1:
The PM2.5 Sampling Sites in North Carolina.

Site Names	Time Period of Sampling	# of Points	Site Type	Kind of Sample
Fayetteville	Jan 2002 - Jan 2004	124	Urban	Speciated Fine PM Conc.
Goldsboro	Jan 2001 - Dec 2003	362	Rural	Fine PM Concentrations
Jacksonville	Jan 2001 - Dec 2003	354	Coastal	Fine PM Concentrations
Kenansville	Jan 2001 - Dec 2003	361	Rural	Fine PM Concentrations
Kinston	Jan 2002 - Jan 2004	123	Rural	Speciated Fine PM Conc.
Kinston	Jan 2001 - Dec 2003	360	Rural	Fine PM Concentrations
Raleigh	Jan 2002 - Jan 2004	146	Urban	Speciated Fine PM Conc.
Raleigh	Jan 2001 - Dec 2003	1084	Urban	Fine PM Concentrations
Wilmington	Jan 2001 - Dec 2003	348	Coastal	Fine PM Concentrations

The primary objective of this study is to investigate the effect of increased ammonia emissions on the PM2.5 concentrations throughout eastern North Carolina. The source of these increased ammonia emissions is the presence of the hog industry. The work conducted here includes analysis of the constituents of PM2.5, their correlations with meteorological variables, and the impact of the hog facilities on PM2.5 concentrations. Another objective is to test how well ISORROPIA can predict the PM2.5 concentrations and under what ambient conditions the model has its best performance in reproducing PM2.5 concentrations.

The model is set for a forward problem, in which the total (both gas and aerosol) concentrations of ammonium, sulfate, sodium, chloride, and nitrate concentrations in addition to relative humidity (RH) and temperature (T) are used to calculate the total aerosol mass. Also, the model is set to run in the thermodynamically-stable state (i.e., solids can be formed when RH decreases below its deliquescence relative humidity (DRH)) instead of the metastable state (i.e., aerosols are in liquid even when RH < DRH). The initial conditions for the ISORROPIA model simulations are listed in Table 2. These conditions were selected based on available observational data in North Carolina and literature values when observational data was not available.

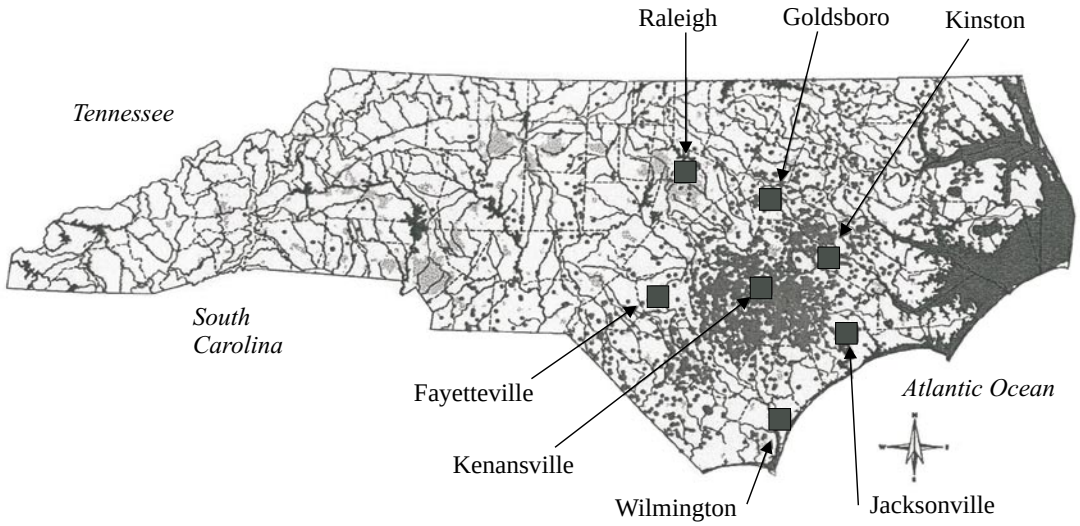

Figure 1:
Map of Hog Facilities in North Carolina and Fine Particulate Sampling Sites

Table 2:
The initial species concentrations and meteorological conditions for ISORROPIA simulations

Input Variables[a]		Kinston	Fayetteville	Raleigh
Sodium		0.22	0.19	0.17
Sulfate	Median	3.43	3.53	3.36
	Minimum	0.58	0.75	0.72
	Maximum	14.30	12.9	13.8
Ammonium	Median	3.13[b]	3.19[c]	5.10[c]
	Minimum	0.32	0.07	0.83
	Maximum	11.50	11.4	16.4
Nitrate	Median	1.07[b]	1.41[c]	1.60[c]
	Minimum	0.26	0.18	0.19
	Maximum	5.24	12.3	19.4
Chloride	Median	0.14[b]	0.33[c]	0.34[c]
	Minimum	0.10	0.02	0.02
	Maximum	0.97	3.20	4.75
Relative Humidity	Median	77	74	74
	Minimum	46	38	36
	Maximum	97	100	98
Temperature	Median	291.00	291.39	289.86
	Minimum	269.61	271.22	268.72
	Maximum	302.44	304.39	301.94

[a] All concentrations are given in µg m^{-3}

[b] (Walker J. T. et al. 2004)

[c] (Bari A. et al. 2003)

For each modeled site (i.e., Kinston, Fayetteville, and Raleigh), three levels of initial total PM species were used: median, minimum and maximum, representing the median, lower and upper limits of the 2002 observations respectively. For each concentration level, the model was run under three meteorological conditions: median RH/median T, minimum RH/maximum T, and maximum RH/minimum T. The output variables include concentrations of gaseous species (i.e., ammonia, hydrochloric acid, and nitric acid) and aerosol species (i.e., sulfate, ammonium, nitrate, sodium, chloride, and water), as well as the pH value.

Observed PM2.5 and its Correlations with Meteorological Variables

The particulate data was first analyzed for its main constituents at the three sites with detailed speciated PM2.5 data over the entirety of the sampling period, as shown in Figure 2. The plot shows the major constituents of PM2.5 to be organic carbon (OC), sulfate, and ammonium consistent with the results by Harrison R. M. et al. (2004), and Tanner R. L. et al. (2004). The additional components of PM2.5 include nitrate, elemental carbon (EC), and over fifty trace elemental species. The PM2.5 OC concentrations were higher in the urban areas, due to large local emissions of primary OC and volatile organic compounds (VOCs). The sulfate and ammonium emissions were found to be larger in the rural site, due to the influence of the hog farming facilities in the rural area.

Figure 3 shows the scatter plots of PM2.5 concentration vs. RH at Raleigh, Kinston, and Wilmington that represent urban, rural, and coastal areas. High PM2.5 concentrations (> 20 µg/m^3) occur with the range of RH between 60 and 90 %, and this effect is more prominent in the urban areas. This trend supports the fact that the overall RH increases the film of water formed on the surface of the particles favors the formation of PM2.5.

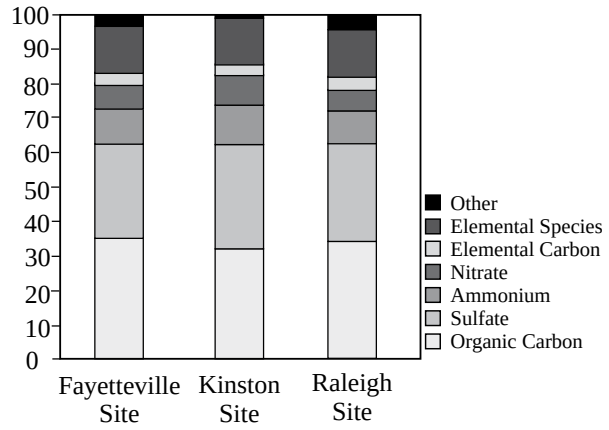

Figure 2:

PM2.5 Composition at Three Speciated Sites (Kinston, Fayetteville, Raleigh) over entire sampling period

Figure 4 shows the correlation between PM2.5 concentrations and wind speeds at three sites. The observed anti-correlation between PM concentration and wind speed is consistent with that of Chu S.-H. et al. (2004) and de Hartog J. J. et al. (2005). The PM2.5-temperature plots for Raleigh, Kenansville, and Wilmington are shown in Figure 5 to represent urban, rural, and coastal areas respectively. Many high PM2.5 concentrations occurred at high temperatures. The slopes range from 0.08 to 0.18 at the urban and the rural sites and 0.01 to 0.02 at the coastal site. To investigate the impact of ammonia on PM2.5 concentrations, the ammonium concentrations were plotted against the total fine particulate concentrations (figure not shown). The values for the slope, intercept, and the coefficient of determination are shown in table 3. There are significant correlations in the two urban sites (i.e., Raleigh and Fayetteville), but no correlation at the rural site (i.e., Kinston). The very low R^2 value in the Kinston correlation plot is due to the local variability of local primary OC PM2.5 emissions (i.e., local biomass burning from farming practices).

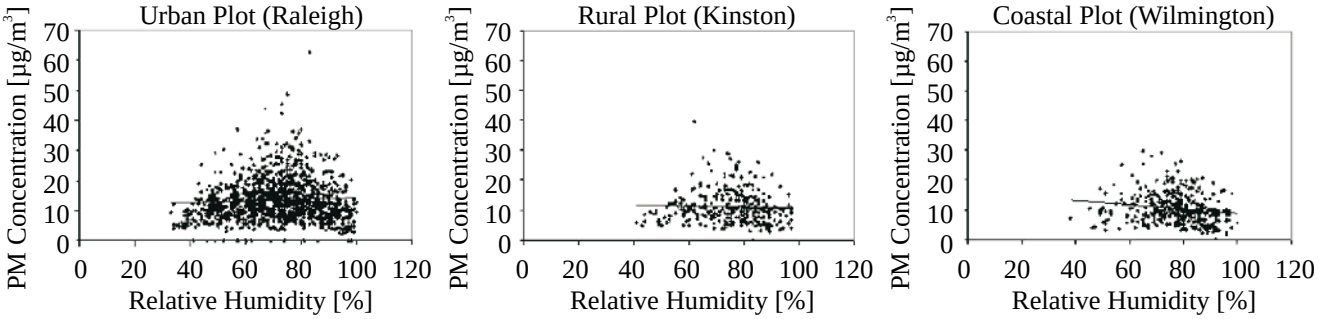

Figure 3:

(a) Urban (Raleigh) Relative Humidity vs. PM2.5 Concentration

(b) Rural (Kinston) Relative Humidity vs. PM2.5 Concentration

(c) Coastal (Wilmington) Relative Humidity vs. PM2.5 Concentration

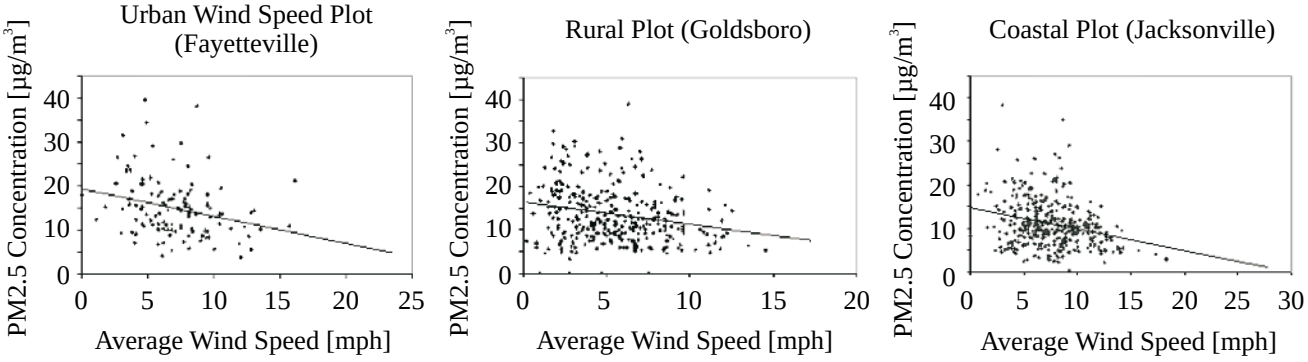

Figure 4:

(a) Urban (Fayetteville) Wind Speed vs. PM2.5 Concentration

(b) Rural (Goldsboro) Wind Speed vs. PM2.5 Concentration

(c) Coastal (Jacksonville) Wind Speed vs. PM2.5 Concentration

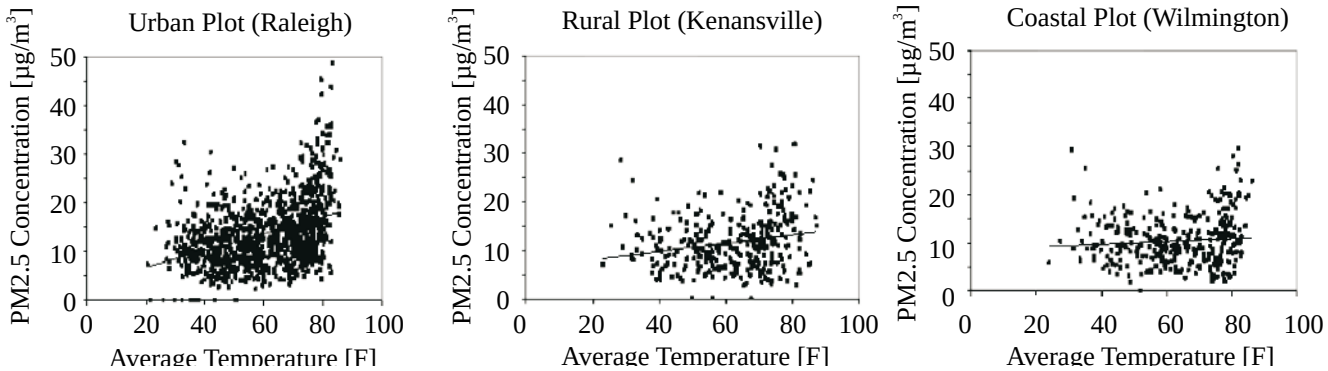

Figure 5:

(a) Urban (Raleigh) Temperature vs. PM2.5 Concentration

(b) Rural (Kenansville) Temperature vs. PM2.5 Concentration

(c) Coastal (Wilmington) Temperature vs. PM2.5 Concentration

Table 3:

The slope, y-intercept, and linear fit R^2 value from the Total PM2.5 vs. Ammonium PM2.5 Plots for Kinston, Fayetteville, and Raleigh

	Slope	Intercept	R^2
Fayetteville	0.0841	0.1758	0.591
Kinston	0.0168	1.21	0.011
Raleigh	0.0995	-0.05	0.712

To investigate the correlation between wind direction and PM distributions, a box whisker plot is made for all seven sites with respect to the eight cardinal directions, as shown in Figure 6. The minimum, 25th percentile, average, 75th percentile, and the maximum of each distribution are plotted. The impact of the hog facilities on PM2.5 concentrations can be seen at some sites. For example, higher PM2.5 average concentrations were found from a southeast flow at Raleigh (urban), which corresponds to Raleigh's orientation to the hog facilities. High PM2.5 concentrations at Kinston (rural) were from the southwest and west directions, which correspond exactly to Kinston's orientation to the majority of hog facilities. The highest average concentrations at Fayetteville were found from the southeast direction, rather than the east from which the emissions of hog facilities come. The weak correlation between the PM2.5 concentrations and the east wind direction at Fayetteville is likely due to the fact that fewer measurements were available at this site and the easterlies were not the prevailing winds during those days with observations. At the other two rural sites (i.e., Goldsboro and Kenansville), relatively homogeneous correlation between PM2.5 concentrations and cardinal directions was found. High PM2.5 average concentrations at Goldsboro were from the southeast, southwest, west, and north directions with the peak concentrations coming from the southeast. The PM2.5 concentrations range from 2.7 to 31.4 with an average of 10.8 µg m^{-3} at Kenansville , which are very high for a small rural town. This indicates the impact of the hog facilities. The two coastal sites (i.e., Jacksonville and Wilmington) have higher concentrations from the southwest and west directions, indicating the impact of emissions from the state of South Carolina. High correlation was also found for the east direction at Jacksonville and the northwest direction at Wilmington.

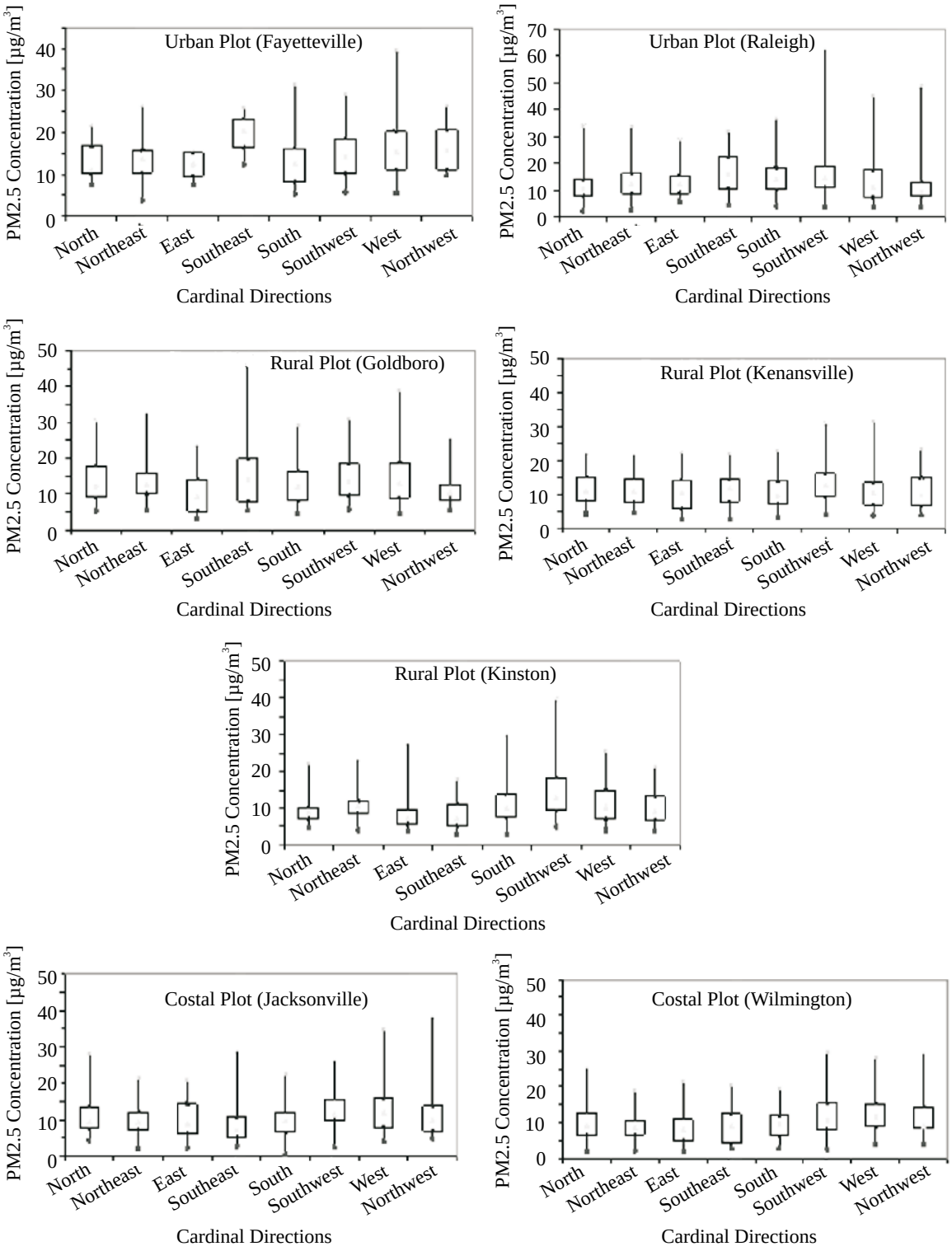

Figure 6:

(a) Urban (Fayetteville) Wind Direction Box-Whisker Plot
(b) Urban (Raleigh) Wind Direction Box-Whisker Plot
(c) Rural (Goldsboro) Wind Direction Box-Whisker Plot
(d) Rural (Kenansville) Wind Direction Box-Whisker Plot
(e) Rural (Kinston) Wind Direction Box-Whisker Plot
(f) Coastal (Jacksonville) Wind Direction Box-Whisker Plot
(g) Coastal (Wilmington) Wind Direction Box-Whisker Plot

Fine Particulate Modeling Results

Figure 7 shows the observed and predicted average total inorganic $PM_{2.5}$ concentrations and its composition at three sites: Fayetteville, Kinston, and Raleigh ("total inorganic PM2.5 or total inorganic PM" is defined as the sum of the four major inorganic constituents: ammonium, chloride, nitrate, and sulfate).

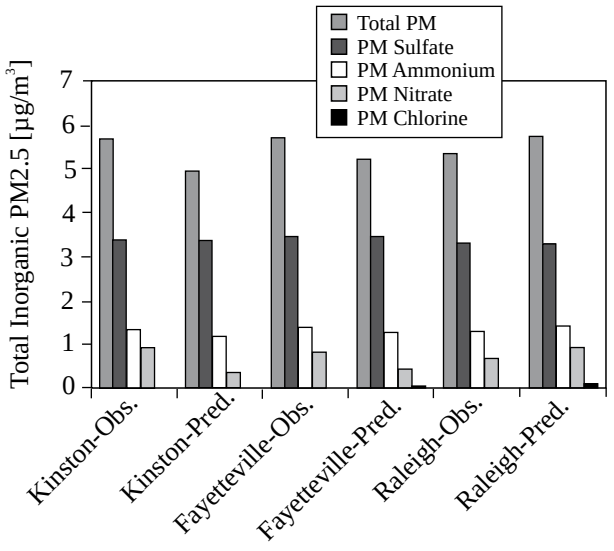

Figure 7:
Observed and Predicted Total Inorganic PM2.5 Concentrations at Kinston, Fayetteville, and Raleigh, NC under median RH and temperature conditions

The predicted values were obtained under the conditions with median initial total PM species concentration, median RH and median T, as shown in Table 2. The observations at all three sites show that sulfate has the largest contribution (approximately 2/3 of the total observed inorganic aerosol), followed respectively by ammonium, nitrate, and chloride. The simulation results from ISORROPIA generally agree well with observed PM2.5 in terms of both magnitude and composition. Compared with observed total inorganic PM2.5 concentration, ISORROPIA underestimates by 0.50-0.75 µg m^{-3} (8.7-12.5 %) at Fayetteville and Kinston, and overestimates the observed values by 0.37 µg m^{-3} (6.9 %) at Raleigh. At all three sites, sulfate has the largest contribution followed respectively by ammonium, nitrate, and chloride. The ammonium concentration at Kinston and Fayetteville is underpredicted by ~0.1 µg m^{-3} (~7.4 %) and that at Raleigh is overpredicted by the same value (7.7 %). The largest differences between observed and predicted values are in the nitrate concentration. It is underpredicted by 0.39 µg m^{-3} at Fayetteville and 0.54 µg m^{-3} at Kinston (48 % and 59 % respectively). The nitrate concentration predicted at Raleigh is 0.25 µg m^{-3} (37 %) greater than the observed nitrate concentrations. The observed chlorine concentrations are nearly zero while the predicted chlorine concentrations at the three sites are < 0.1 µg m^{-3}. At each site, the predicted pH and aerosol water concentrations are in the range of 7.53-7.56, and 5 µg m^{-3} respectively. The model gives the best agreement against observations at Raleigh among the three sites.

Figure 8 shows the predicted total inorganic PM2.5 concentration at the maximum initial pollutant concentrations at each site under the three different meteorological settings.

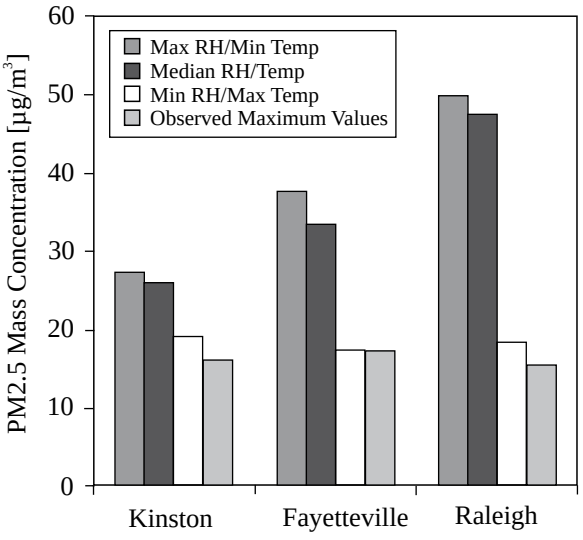

Figure 8:
Predicted Total Inorganic PM2.5 Concentrations under three meteorological conditions and maximum observed Total Inorganic PM2.5 Concentrations at Kinston, Fayetteville, and Raleigh, NC

The maximum observed values are also plotted for comparison. For the median RH/median T and the maximum RH/minimum T conditions, the predicted total PM2.5 inorganic aerosol concentrations range from 26 to 50 µg m^{-3} at the three sites, which consistently overpredicts the observed maximum concentrations (15-18 µg m^{-3}) at all sites**.** The predicted total inorganic PM2.5 concentration increases as the urban development of the area increases (Kinston (rural), Fayetteville (small city), Raleigh (large city)). These differences are due to differences in the predicted particulate nitrate concentration, which is factors 2 and 3 higher at Fayetteville and Raleigh, respectively, than that at Kinston. The predicted particulate ammonium concentration is higher by 32 % and 80 % at Fayetteville and Raleigh, respectively, due to formation of ammonium nitrate. With the higher sulfate concentrations at Kinston and Raleigh, the aerosol is much more acidic at these sites (with pH values of 4.5-4.8), whereas that at Fayetteville is more neutral (6.8). The predicted total inorganic aerosol concentrations range from 17.23 to 19.09 µg m^{-3} at the three sites under

the minimum RH/maximum T condition. Such a condition favors evaporation of nitrate and water, resulting in zero nitrate and water concentration in the aerosol phase. The aerosol consists of primarily ammonium sulfate salt. The differences in predicted total inorganic aerosol concentrations among these sites are thus much smaller.

A similar plot is shown at the minimum pollutant concentrations at each site in Figure 9.

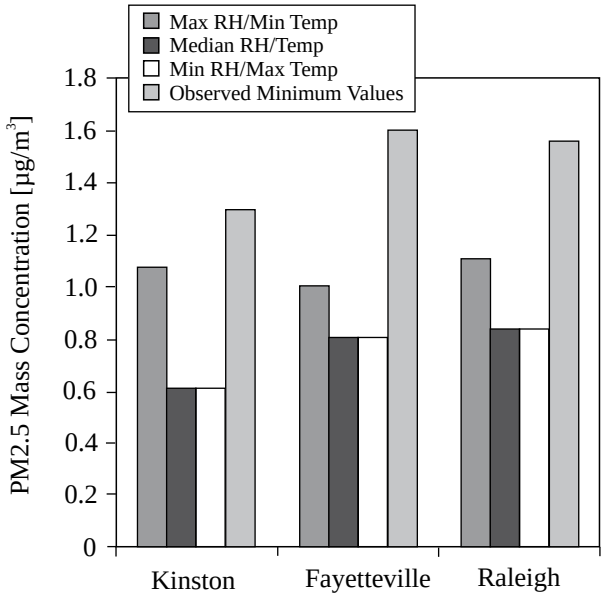

Figure 9:
Predicted Total Inorganic PM2.5 Concentrations under three meteorological conditions and minimum observed Total Inorganic PM2.5 Concentrations at Kinston, Fayetteville, and Raleigh, NC

The model underestimates the observed minimum concentrations by less than 1 µg m^{-3} at each site. Under the median RH/median temperature and the minimum RH/maximum T conditions, the total inorganic PM2.5 concentrations are the same and they consist of sulfate salts only. The nitrate concentrations are either zero or negligible. Under the maximum RH/minimum T conditions, some nitrate forms. The total PM species concentrations predicted at the three sites range from 1.01 to 1.11 µg m^{-3}. Under this condition, the urban areas are characterized by 20 % more sulfate than the rural site, but the rural site (i.e. Kinston) had slightly more nitrate, ammonium, and chloride resulting in total PM2.5 concentration that is slightly higher than that at Fayetteville but lower than that at Raleigh.

Conclusion

The concentrations and trends of PM2.5 in eastern North Carolina are studied with data analysis and an aerosol thermodynamic box model that predicts the gas/particle partitioning of PM. The unique emission fluxes of pollutants (e.g. ammonia) from the hog industry and their impacts on PM concentrations make this region a unique environment to understand the role of these emissions in PM formation. The major constituents of fine PM2.5 from the greatest to the least are OC, sulfate, ammonium, nitrate, and EC. Higher PM2.5 concentrations tend to occur between 60 and 90 % RH with this effect being more pronounced in urban areas. There is a positive relationship between temperature and PM2.5 concentrations, and a negative relationship between wind speed and PM2.5 concentrations. The box-whisker plots of wind direction demonstrate that there is a connection between hog facility density and fine particulate concentration, but with the limited data, these concentrations could not be attributed to any specific pollutant.

ISORROPIA is used to simulate the gas/particle partitioning and the total inorganic aerosol concentration at three sites in eastern North Carolina. The model predictions show that the major predicted constituents of inorganic aerosols are sulfate, ammonium, and nitrate, which agrees with the overall measurements. The predicted average total inorganic concentrations are slightly (< 1 µg m^{-3}) lower than the observations. While the model predicts the concentrations of sulfate and ammonium that are in good agreement with observations, it tends to underpredict the observed particulate nitrate concentrations by 0.22 µg m^{-3} (27.5 %) at all three sites. The simulation results are sensitive to initial total PM concentrations and meteorological conditions, with the highest secondary PM formation occurring under the condition with maximum initial total PM concentrations, maximum RH, and minimum temperature.

Acknowledgements

This research was sponsored by a research initiative grant from the United States Department of Agriculture. YZ's time is supported by the NSF CAREER award No Atm-0348819. SG thanks Ryan Boyles of the North Carolina State Climate Office, and Hoke Kimball and Wayne Cornelius from the North Carolina Division of Air Quality for their assistance in obtaining the data. Thanks are also due to Dr. Athanasios Nenes from the Georgia Institute of Technology for providing the source code of ISORROPIA. (The latest version of the ISORROPIA code and more information about the code may be obtained at http://nenes.eas.gatech.edu/ISORROPIA).

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Influence of soil type and soil moisture on PM emissions from soils during tillage

R. Funk[1], W. Engel[1], C. Hoffmann[1], and H. Reuter[1]

Abstract

Tillage is an important cause of PM emissions from soils. Measurements in rural areas in Germany indicated many times higher fine dust emissions by tillage operations than by wind erosion. The main controlling factor is the soil moisture, or the vertical distribution of moisture in a soil profile at the moment of tillage. As the emission is a result of some parameters which can not be controlled in field experiments, a stepwise analysis of the main influencing factors was chosen. First, a wind tunnel was used as cross-flow gravitational separator to investigate the relation between soil type, soil moisture and PM emission. Twelve soils of different texture (7 sandy, 2 silty, 1 clayey and 2 organic soils) were investigated with regard to their water content, ranging from 0 to 40 mass per cent. The results show that soils can emit dust over a certain range of moisture, but already a small increase in soil moisture causes a distinct reduction of dust emission. The threshold water content for fine dust emission of soils was between 2 to 5 mass per cent for sandy soils, 5 to 10 mass per cent for silty soils, about 30 mass per cent for the clayey soil and 25 to 45 mass per cent for organic soils.

The wind tunnel results were used to calculate the PM10 emission potential of a sandy soil in spring and late summer. In spring only the upper 2.5 cm were dry enough to emit PM10, whereas in summer the soil was desiccated to the entire tillage depth. The calculated PM10 emission potential for a tillage depth of 20 cm resulted in 13.4 g per m² for the soil moisture conditions in spring and 76.8 g per m² in summer.

Our results show the importance of the vertical soil moisture profile on the PM emission of soils during tillage. So, the emission factors resulting from field operations should preferably be related to the affected amount/volume of a soil, which is dry enough to emit PM, than to the affected area.

Keywords: soil moisture, particulate matter, emission, tillage

Introduction

Fine dust particles seldom emit directly from soil surfaces. They usually need a releasing process as wind erosion or tillage operations (Gillette D. 1977, Green F. H. et al. 1990, Clausnitzer H. et al. 1996, Alfaro S. C. 2001, Kjelgaard et al. 2004). Wind erosion is limited to a certain extent because a susceptible soil surface and a given erosivity of the climate have to coincide. It is temporally restricted to the spring months and constrained spatially by the acreages of root crops, corn and summer cereals, which amount to about 20 per cent of the agricultural land area in Northern Germany (Federal Statistical Office Germany 2006).

Dust emission resulting from tillage operations affects all soils, even those which are considered to be non-erodible. This is mainly caused by higher contents of silt and clay particles, which support the formation of aggregates or crusts. On the other hand these soils have a higher potential for dust emission when they are disturbed by the impact of tillage tools. On the North European plains dust emission induced by tillage was measured as being four to six times higher than the dust emission by wind erosion events (Goossens D. et al. 2001, Goossens D. 2004). The correlations between soil tillage and dust emission have been investigated in many studies, ranging from effects on human health to the effects of losses of fine material on soil fertility and air quality (Louhelainen K. et al. 1987, Clausnitzer H. et al. 1996, Nieuwenhuijsen M. J. et al. 1998, Nieuwenhuijsen M. J. Schenker M. B. 1998, Schenker M. B. 2000, Holmen B. A. et al. 2001a/b, Trzepla-Nabaglo K. et al. 2002, Cassel T. et al. 2003, Goossens D. 2004). Most of these studies contain no or only general information about the soil texture and soil water content, so that is not possible to derive the potential of soils as a source for dust emission from these concentration measurements. The main controlling factor is the soil moisture, or more precisely the vertical distribution of moisture in a soil profile at the moment of tillage. Soil moisture is one of the most important factors which limits wind erosion and therefore dust emission as well (Chepil W. S. 1956, Weinan Ch. et al. 1996). The influence of soil moisture on erodibility has been investigated well in empirical or physically-based studies (ChepilW. S. 1956, Bisal F. et al. 1966, Mc Kenna-Neumann C. et al. 1989, Saleh A. et al. 1995, Weinan Ch. et al., 1996, Cornelis W., Gabriels D. 2003, Cornelis W. et

[1] Leibniz-Centre for Agricultural Landscape Research, Institute of Soil Landscape Research, Müncheberg, Germany

al. 2004). In most cases increasing water content results in decreasing wind erosion and dust emissions. Although most of the processes responsible for dust emission of soils are known in detail, there are still some deficits in implementing this knowledge to obtain a more soil-related balance of dust emission caused by tillage. The objective of our study was therefore to derive a soil-related emission factor, which also considers the actual conditions in the field.

Material and Methods

Wind tunnel investigations

The experimental setup in the laboratory was intended to reproduce the basic processes in the field during tillage in a simple and repeatable way. These processes can be described as follows: soil particles will be lifted and accelerated by the action of the tillage tools and the tyres of the tractor into the direction of the operation. Then gravity and the pull of the moving tractor result in a vertical and horizontal component of the separation process depending on size and density of single particles or aggregates and the speed of the tillage tool.

Dust emission measurements took place in the wind tunnel of the Institute of Soil Landscape Research in Müncheberg (Funk R. 2000). The wind tunnel is generally used to investigate wind erosion processes, but it can also be applied as a cross-flow gravitational separator according to standardised particle size analysis by air classification (DIN 66118). A wind tunnel is a suitable tool to carry out a separation in this way because height of fall, sedimentation distance and wind speed can be adjusted to optimise

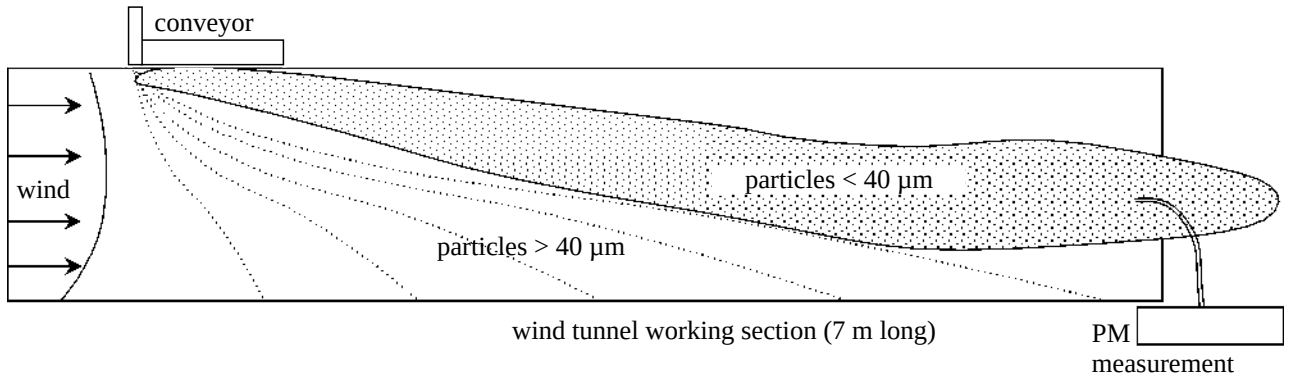

Figure 1:
The experimental setup in the wind tunnel for cross-flow gravitational separation

Table 1:
Soil texture, humus content and water contents of the investigated soils

Site	Code	Sand 2000-63µm %	Silt 63-2µm %	Clay < 2 µm %	Humus %	SWC 60 °C* M%	SWC air-dry** M%
Klockenhagen	KLOC	91.8	7.4	0.8	1.31	0.19	0.61
Siggelkow	SIGG	89.4	8.3	2.3	1.32	0.29	0.66
Gottesgabe	GOGA	87.3	6.9	5.8	1.33	0.15	0.56
Muencheberg	MUEB	82.5	14.1	3.4	0.90	0.23	0.46
Sandhagen	SAHA	81.2	15.7	3.1	1.13	0.21	0.75
Penkow	PENK	73.8	22.4	3.8	1.35	0.25	1.41
Gross Kiesow	GRKI	72.8	24.7	2.5	1.28	0.28	1.04
Hildesheim	HILD	2.1	81.9	16.0	0.94	0.46	1.85
Bad Lauchstedt	BALA	11.0	65.0	24.0		0.75	2.85
Seelow	SEEL	14.3	28.6	57.1	2.18	2.63	4.15
Heinrichswalde	HEIN	74.4	15.0	10.6	23.3	3.33	6.91
Rhinluch	RHIN				40.9	9.86	21.2

* SWC 60 °C – gravimetric soil water content (mass per cent) after 24 hours oven drying at 60 °C

** SWC air-dry – gravimetric soil water content after drying in the laboratory (21°C, 60% rH)

the separation process for certain particle sizes. The working section of our wind tunnel has a length of 7 m and a cross section area of 0.7 x 0.7 m. The wind speed in the centre of the tunnel was set to 3 m/s. In contrast to regular tests in the wind tunnel we minimised the boundary layer below 10 cm height and adjusted a more laminar flow.

The soil material was supplied by a conveyor which was placed at the beginning of the working section on top of the wind tunnel (figure 1). A plate, 0.5 cm thick and with a cut-out of 10 x 20 cm, was placed at the conveyor belt. Cut-out and thickness of the plate result in a volume of 100 cm³ which was filled with the soil material, smoothed and covered by a plastic plate to minimise moisture losses during the runs. After starting the conveyor the soil material fell off through a 10 cm wide slot into the working section at a constant rate in 6 minutes.

The soils were taken from the plough-horizon of seven sandy soils, two silt loam soils, two organic soils and one clay soil. The soils were air-dried in an air-conditioned laboratory (temperature: 21 °C, relative humidity: 60 %) and sieved for the fraction less than 2 mm. One part of each soil was dried at 105 °C to obtain the amount of hygroscopic water which had remained in the air-dried soil. Samples of 300 g each was moistened with distilled water. Soil water contents of the following gradations were set: 105 °C dried, 60 °C dried, air-dried and depending on the texture in 6 to 10 further steps of about a half mass per cent up to a distinctly visible moist condition of the sample. The samples were stored in hermetically sealed Erlenmeyer flasks for 24 hours. The next day the soil samples were placed on the conveyor, covered and supplied to the wind tunnel.

The dust fraction was measured with a dust monitor (Grimm #107 Spectrometer), which continuously detects all particles between 0.3 to 30 µm and registers these in three classes as particle mass PM10, PM2.5 and PM1.0 in µg per m³. The threshold water content for dust emission was appointed when, compared with the base load in the wind tunnel, no increase in the PM10 concentration could be measured. Multiple regression calculations were performed using WinStat software (R. Fitch Software).

Field measurements

The field measurements were conducted on loamy sand in Muencheberg (Brandenburg, Germany), which was ploughed under typical moisture conditions, such as in spring and in summer. The field measured 50 m x 50 m. Before tillage, the vertical soil moisture profile was measured in steps of 1 cm at several points by a near Infrared-Reflexion-Photometer (Pier-Electronic GmbH).

For ploughing we used a 100 kW tractor with a 3-share plough and a working width of 1.25 m and a working depth of 0.2 m. The tillage direction was perpendicular to the wind direction (figure 2). The dust monitor (GRIMM #107) was positioned in combination with a meteorological station two meters away from the leeward field boundary with the air inlet at a height of 2 meters. PM10, PM2.5 and PM1.0 were measured every 6 seconds simultaneously with wind speed (cup anemometers in two heights, 2 m and 0.5 m), wind direction, temperature and relative humidity. The aim was to measure only that dust which leaves the field and to get a horizontal intersection of the dust cloud, which was obtained by the repeated passage of the tractor and the increasing distance with any passage.

A Lagrangian dispersion model GRAL (Graz Lagrangian Model) was used to obtain PM area related emission factors from the concentration measurements. GRAL is a well-validated short range numerical model and applicable to a wide range of wind speeds and atmospheric stabilities (Oettl D. et al. 2001). Emission factors of PM were obtained by modelling the dispersion from the test field, treating it as an area source. In the simulations it was assumed that the PM emissions are initially mixed up to 2 m above ground level due to the tractor-induced turbulence (Oettl D. et al. 2005).

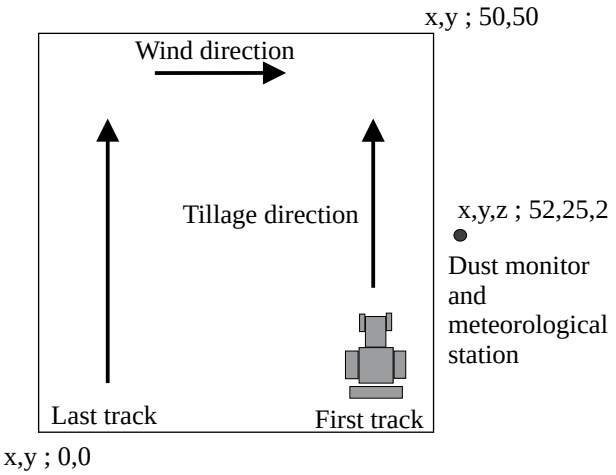

Figure 2:
Sketch of the field measurements of tillage induced PM emissions

Results

Wind tunnel investigations

Our results show that soils can emit particulate matter over a certain range of moisture. Great differences in the PM10 emission were already measured between the 105°C-, 60°C- and air-dried soil samples (figure 3). The water content of the 105°C-dried samples was assumed to be zero, the water contents of the 60°C- and air-dried samples are listed in table 1. Sandy and silt soils had the highest fine dust emission rates of both oven-dried samples and

the lowest of the air-dried sample. The clay soil does not show such a difference. The PM10 emission of the oven-dried samples resulted in: sandy > silty > clay, whereas the emission of the air dried samples was: sandy < silty < clay. This opposing trend can be explained by the small contact areas between the particles in the sand and clay fraction in sandy soils, which result in weak bonding forces mostly caused by the adsorptive water between the contact points. Water films resulting from molecular adsorption only appear on particles of the clay size, which are attached to the outside of the sand and silt grains and form a large surface for evaporation (figure 4).

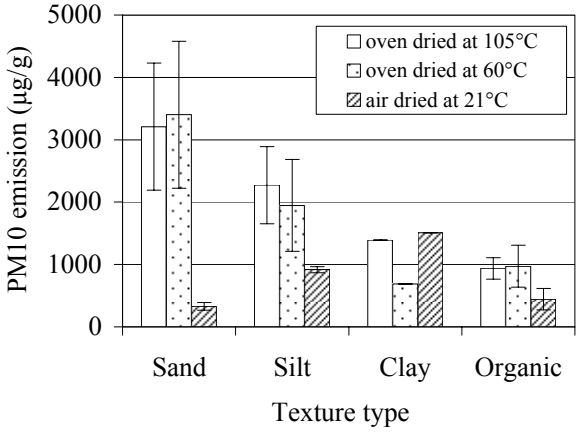

Figure 3:
Dust emission of sandy (n = 7), organic (n = 2), silty (n = 2) and clay (n = 1) soils using different drying intensities (oven dried at 105°C and 60°C, air dried at 21°C)

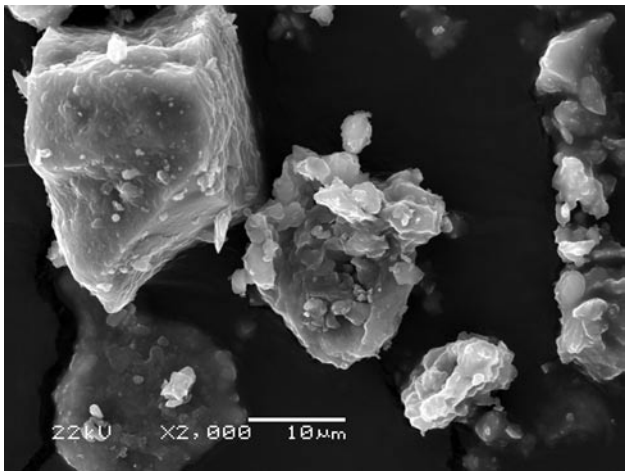

Figure 4:
Scanning electron microscopy of a mineral dust sample

Figure 5 shows the relationship between the PM10 emissions of all investigated soils to the content of particles of this size in the soil. These are all particles in the clay and fine silt fraction (< 6.3 µm in diameter) according to the German Standard for soil texture classification, which are nearest to the PM10 size. The PM10 emission potential of a soil is closely related to the content of these particle sizes.

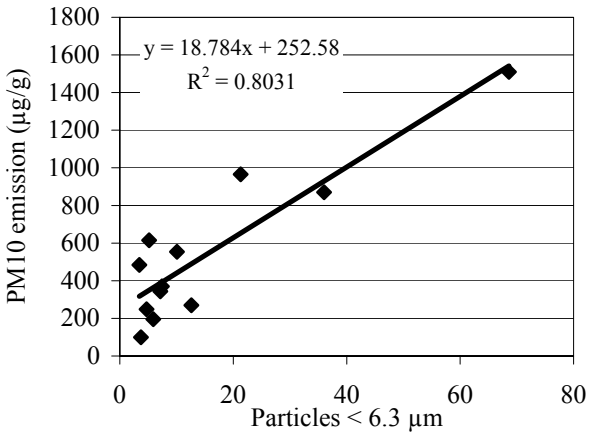

Figure 5:
Relationship between the content of particles < 6.3 µm (clay and fine silt) and the PM10 emission of the air dried samples (all investigated soils)

The relationship between the PM10 emission and the increasing gravimetric water content of the sandy soils is shown in figure 6, that of silt, clay and organic soils in figure 7. The results show the reduction of dust emission with increasing soil moisture. Even small differences in soil moisture caused distinct changes in dust emission, resulting in an exponential curve progression for all soil types. In an attempt to relate the PM emission to soil moisture and parameters of the soil texture and humus content, a multiple regression analysis was performed. Table 2 summarizes the coefficients of the regression equations, which have effected the best r^2. Above a certain water content no dust was emitted. This can be regarded as the texture related threshold of soil moisture. These threshold values of soil moisture are between 2 to 5 % for sandy soils, 5 to 10 % for silty soils, up to 20 % for the clay soil and 25 to 45 % for organic soils. The share of PM2.5 amounts to about 6 per cent, the portion of PM1.0 to about 1 per cent of the PM10 mass. These relations were relatively constant for all investigated soils and did not change with increasing soil moisture.

Table 2.
Parameter of multiple linear regressions of the form:
ln PM (µgm^{-3}) = a + b SWC (M%) + c silt (%) + d clay (%) + e humus (%), Significance level p = 0.05

Soil textural class		a	b	c	d	e	r²
Sand	ln PM10	7.07	-1.182	0.115		-1.73	0.77
	ln PM2.5	5.35	-0.980	0.070		-2.35	0.54
	ln PM1.0	4.24	-0.955	0.054		-2.48	0.42
Silt + clay	ln PM10	4.95	-0.248		0.068		0.56
	ln PM2.5	2.10	-0.347		0.078		0.55
	ln PM1.0	1.22	-0.363		0.067		0.70
Organic soils	ln PM10	11.32	-0.117			-0.095	0.86
	ln PM2.5	9.67	-0.159			-0.125	0.87
	ln PM1.0	5.03	-0.145			-0.052	0.41

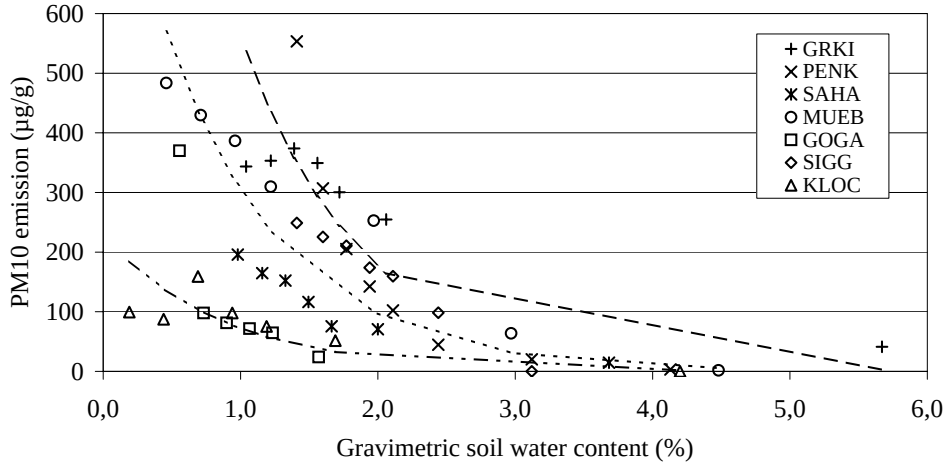

Figure 6:
PM10 emission of all sandy soils, curves are calculated with multiple regression (see table 2), shown curves are examples of GRKI (72.8% sand), MUEB (82.5% sand) and KLOC (91.8% sand)

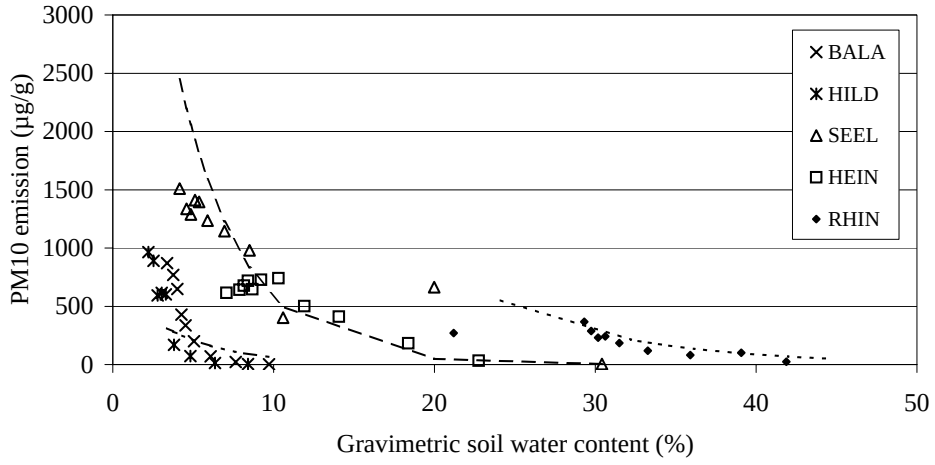

Figure 7:
PM10 emission of all silt, clay and organic soils, curves are calculated with multiple regression (see table 2), shown curves are examples of RHIN (organic soil), SEEL (clay soil) and BALA (silt soil)

Relevance for the derivation of emission factors

The relevance of our results for the derivation of emission factors is demonstrated by field measurements on a sandy soil in Muencheberg, which was ploughed in spring and in summer. The soil texture is given in table 1. The soil moisture of the first centimetre was approximately the same; only the soil moisture depth profile differed at both times and is shown in figure 8.

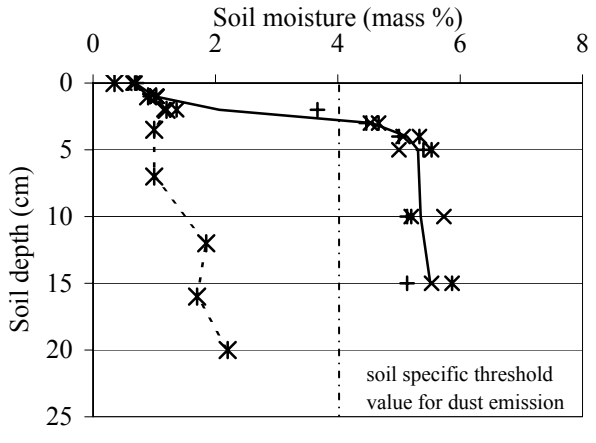

Figure 8:
Soil moisture depth profiles at two dates of ploughing

The soil specific threshold value of moisture for dust emission was estimated as about 4 %. Under the moisture conditions in spring only the uppermost 2.5 cm of the soil are dry enough to contribute to the emission of dust particles. The conditions in summer are characterised by soil moisture below the threshold value for the entire ploughing depth. We derived a PM10 emission potential (EP_{PM10}) of 13.4 g per m² (± 0.8 g) in spring and 76.8 g per m² (± 4.6 g) in summer for a ploughing depth of 20 cm, using the above determined soil moisture – dust emission relationship in steps of 1 cm.

$$EP_{PM10} = \sum_{i=1}^{20} TDE_i \cdot \rho \cdot V$$

with
- EP_{PM10} - PM10 emission potential (µg per m²)
- TDE - total dust emission of the soil related to the water content (µg per g)
- ρ - bulk density of the soil (g per cm³)
- V - volume of the layer i per m² (10,000 cm³ per m²)

PM measurements and subsequent modelling with the dispersion model GRAL resulted in an emission of 0.12 g per m² of the moist soil in spring and 1.05 g per m² of the dry soil in summer (Oettl et al. 2005). The same tillage operation at the same soil resulted in about 9-times higher emissions in summer mainly induced by the lack of soil moisture. There is a close relationship to the affected volume or mass of the soil contributing to the dust emission, which is 8-times higher in summer (20 cm in summer, 2.5 cm in spring).

Conclusion

Dust emission from tillage operations are closely related to the soil moisture conditions. This aspect has not been considered in previous studies so far. In this study we followed a stepwise analysis of the main influencing factors of dust emission from tillage operations. Firstly, the basic relations between soil texture, humus content and soil water content were investigated by using a wind tunnel as cross-flow gravitational separator. It was possible to show that soils can emit PM over a certain range of moisture. The relationships of soil water content, soil texture and humus content on PM10, PM2.5 and PM1.0 emissions could be derived. Threshold values of soil moisture for PM emission were determined which ranged between 2 to 5 % for sandy soils, 5 to 10 % for silty soils, up to 20 % for the clay soil and 25 to 45 % for organic soils.

Applying these findings on moisture conditions in spring and in summer resulted in good correlations between the derived PM emission potentials of a soil and measured emissions while ploughing. Our results show the necessity of considering the soil water content and its vertical profile for the derivation of emission factors. So, the emission factors of field operations should be related to the affected amount/volume of a soil, which is dry enough to emit PM, rather than to the affected area.

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A methodology to constrain the potential source strength of various soil dust sources contributing to atmospheric PM10 concentrations

H. Denier van der Gon[1], M. Schaap[1], A. Visschedijk[1], and E. Hendriks[1]

Abstract

Crustal material or soil particles typically make up 5 - 20 % of the mass of ambient PM10 samples. In certain regions and/or specific meteorological conditions the contribution may even be higher. Crustal material may originate from distinctly different sources e.g., wind erosion of bare soils, agricultural land management, driving on unpaved roads, resuspension of road dust, road wear, handling of materials and building and construction activities. Despite the importance of crustal material in total PM10 mass, the sources are still poorly understood and not well-represented in emission inventories. This is due to the fact that some sources can be defined as natural sources (e.g., erosion) whilst others like re-suspension are not recognized as primary emission but a re-emission. Separating the source contributions is difficult because the unique tracers for crustal material in PM10 samples (e.g., Si, Ti) do not allow a distinction between the potential sources of this material. To make progress in our understanding of the crustal material source strengths we need a combination of flux measurements, emission estimates, chemical analysis of ambient PM10 samples and atmospheric transport modelling. In this paper we present a methodology to check first order estimates of the various source strengths. Simple and therefore transparent emission functions are combined with activity data or land use maps to make emission grids. The gridded emissions are used as input for the LOTOS-EUROS model to calculate the resulting concentrations. The predicted concentrations will be compared with observations in various parts of Europe derived from the literature. This will give an indication if the source strengths are in the right order of magnitude to explain observed crustal material contributions to ambient PM10. Next, by varying the source strengths of the individual categories and implementation of a meteorological dependency we will investigate if the patterns of the predicted concentrations are in line with observations.

Keywords: particulate matter, soil particles, resuspension, wind ersosion, crustal material, emissions, air quality

1 The relevance, chemical composition and sources of crustal material.

Particulate matter (PM) is the generic term used for a type of air pollution that consists of complex and varying mixtures of particles suspended in the atmosphere that has been found to present a serious danger to human health. Particulate pollution comes from such diverse sources as coal or oil fired power plants, vehicle exhaust, wood burning, mining, and agriculture. Airborne PM includes many different chemical constituents. PM10 samples collected on a filter can be analyzed for chemical composition. Sources contributing to the PM10 concentrations may emit particles with a unique chemical composition that are enriched, relative to particles from other sources, in certain elements. For example, vanadium and nickel have typically been used as tracers for emissions from fuel oil combustion. Based on the concentrations of the tracers an estimate of the total amount of PM10 emitted by the particular source can be made. Crustal material (mineral matter, soil particles) typically makes up 5 - 20 % of the mass of ambient PM10 (table 1).

In certain regions and/or specific meteorological conditions the contribution may be higher. This is illustrated in table 1 with the values for Northern EU and Southern EU where the contribution is elevated due to the use of studded tires and desert dust, respectively.

Crustal material may originate from distinctly different sources e.g., wind erosion of bare soils, agricultural land management, driving on unpaved roads, resuspension of road dust, road wear, handling of materials and building and construction activities (figure 1). Despite the importance of crustal material in total PM10 mass, the sources are still poorly understood as well as their relative contribution to total crustal PM10. In this paper we present a methodology to check first order estimates of the various source strengths (figure 1). Simple, transparent emission functions are combined with activity data or land use maps to make emission grids.

[1] TNO Built Environment and Geosciences, Business unit Environment, Health and Safety, Apeldoorn, The Netherlands

Table 1:
Mean annual levels (µgm^{-3}) of PM10, PM2.5, mineral elements, and the equivalent contributions to bulk mass concentrations (% wt), recorded at regional background (RB), urban background (UB) and kerbside stations (RS) in Central EU (examples from Australia, Berlin, Switzerland, The Netherlands, UK), Northern EU (13 sites in Sweden) and Southern EU (10 sites in Spain).

	Central EU			Northern EU)			Southern EU		
	RB	UB	RS	RB	UB	RS	RB	UB	RS
PM10 (µgm^{-3})	14 – 24	24 – 38	30 – 53	8 – 16	17 – 23	26 – 51	14 – 21	31 – 42	45 – 55
Mineral matter (µgm^{-3})	1 – 2	3 – 5	4 – 8	2 – 4	7 – 9	17 – 36	4 – 8	8 – 12	10 – 18
% Mineral matter PM10	5 – 10	10 – 15	12 – 15	20 – 30	35 – 45	65 – 70	12 – 40	25 – 30	25 – 37
PM2.5 (µgm^{-3})	12 – 20	16 – 30	22 – 39	7 – 13	8 – 15	13 – 19	12 – 16	19 – 25	28 – 35
Mineral matter (µgm^{-3})	0.5 – 2	0.4 – 2	1 – 2	1 – 3	2 – 4	4 – 6	1 – 3	2 – 5	4 – 6
% Mineral matter PM2.5	2 – 8	2 – 8	5	15 – 25	25 – 30	30 – 40	8 – 20	10 – 20	10 – 15

Source: Querol X. et al (2004)

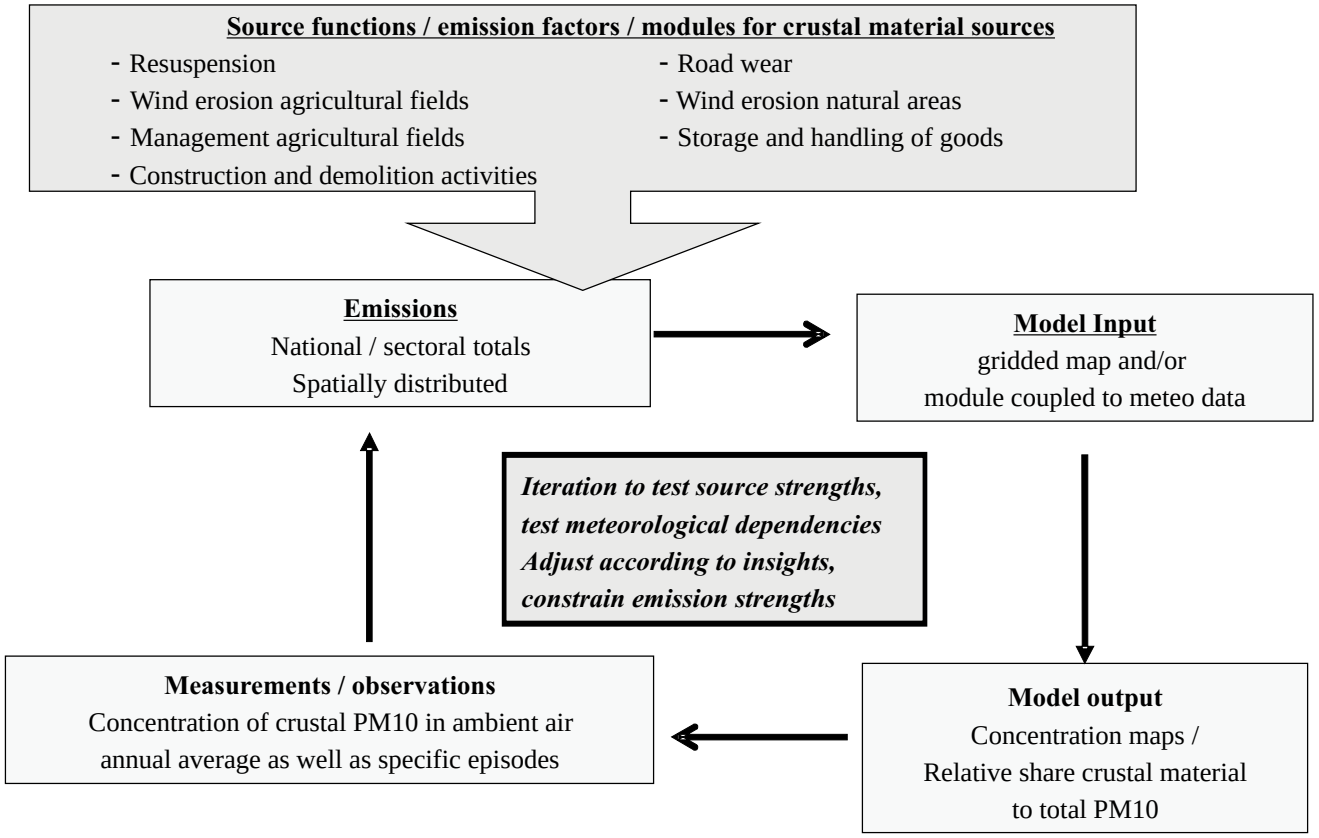

Figure 1:
Outline of the methodology to constrain the source strength of various crustal material sources contributing to PM10 using an atmospheric chemistry and transport model and observational data

2 Emission functions and source strengths

Eventually all different sources of crustal material (figure 1) should contribute their own unique emission pattern and strength to the gridded data going into the atmospheric transport model. So, for each of these emission causes an emission estimate and/or module that calculates emission within the model needs to be constructed. To illustrate the methodology we present three examples.

- Windblown dust from bare soils; A wind velocity driven factor is applied to all bare soils following the parameterization of van Loon M. et al. (2005). This is a simplified version of paramterizations developed to predict wind blown dust from deserts. As a first approximation we applied this function to all arable lands according to the CORINE land use map (CORINE, 2003) but at only 1/50 of the source strength because arable lands will have a lower emission than continuous bare soils or deserts. Furthermore we assume no emissions occur on rainy days. Next, sensitivity studies will be done by increasing the bare soil area and adjusting the source function. Obviously our first assumptions may easily be an order of magnitude wrong and comparisons with measurement data in particular regions will be used to tune the source strength.
- Road wear or pavement wear; Pavements consist mainly from rock material and the contribution of pavement wear to the total PM10 emission factor is difficult to separate from the contribution of windblown- or resuspended soil and other geological material. However in a road tunnel the amount of windblown soil dust coming from the shoulders or elsewhere will be limited and a first approximation can be that all the crustal material in PM10 produced in the tunnel originates from road wear. Gillies J. A. et al. (2001) reported that ~12 % of the PM10 in the Sulpeveda tunnel was of geological origin. Denier van der Gon H. A. C. et al. (2003) used this observation in combination with the chemical analysis of PM10 formed in the the Maas tunnel (Rotterdam, The Netherlands) to derive an emission factor of 3 - 4 mg per vehicle kilometre (vkm) for crustal material from road wear. Road wear will be dependent on the pavement material but also on the frequency of braking and cornering causing additional friction. In the tunnel environment this will be limited compared to the average urban traffic. Hence we see the tunnel road wear emission factor as a lower value. In the literature road wear emission factors are often in the range of 8 - 10 mg/vkm which is consistent with 3 - 4 mg/vkm being a lower limit.
- Resuspension by traffic; The material that is being resuspended will vary with the nature of the local circumstances but it is typically likely to include particles from vehicle tyre and brake dusts, primary exhaust emissions that have settled out of the atmosphere (perhaps adhering to larger particles) and environmental dusts from many sources eg pollen, sea salt, construction work and wind blown soils. Furthermore the contribution of resuspension to total PM will be depedendent on the meteorological conditions; during rain and/or while the surfaces are wet the resuspension will be much lower. The mix of particles with varying chemical composition, dependencies on local and climatic conditions make this a difficult source to quantify and generalize. However, it is widely acknowledged that resuspesion is potentially a very important source in the same order of magnitude as traffic exhaust emissions. In our first, explorative approach we take an emission factor of 80 mg/vkm and than selectively switch this off on rainy days.

It should be noted that road wear and resuspension will both be coupled vehicle kilometres driven, hence they cannot be separated. Furthermore, as discussed above, not all resuspension will be crustal material. Therefore the 80 mg/vkm that we use as a first approximation for crustal PM10 from traffic contains only the crustal fraction of resuspension emissions and includes road wear.

3 Spatially distributed emission maps

The basic procedure to come to a high resolution gridded emission map that can be used as input for a model is to redistribute total national sectoral emissions using high resolution distribution patterns, also called proxi data. The TNO proxi data consist of recent geographical distribution data for point sources and area sources and are described by Visschedijk A. H. J. and Denier van der Gon H. A. C. (2005). For example, in the present study the emission maps for crustal material from road wear and resuspension are made by calculating the total amount road wear and/or resuspension from annual amount of kilometres driven by vehicle category (e.g. heavy duty vehicles, light duty vehicles and passenger cars) within a country taken from the CAFE base line scenario (Amman M. et al., 2005) and split in a highway, urban and rural part. For the spatial distribution of the emissions the highway part is distributed according to digitized maps of highways, using traffic intensities by highway section for LDV and HDV from Eurostat (Eurostat, 2003). The rural part of the road transport emission is distributed according to the digital maps of the European rural road network. The urban part of the road transport emissions is distributed according to a high resolution population density map (CIESIN, 2001).

4 Model description

We have used the LOTOS-EUROS model (Schaap M. et al., 2007) to calculate the crustal material concentrations in ambient air over Europe using meteorological data from 1998 and 1999. The LOTOS-EUROS model is a 3D-chemistry transport model developed to study the formation, transport and sinks of oxidants, particulate matter and heavy metals. LOTOS-EUROS is applied for the region that spans from 10 °W to 40 °E and from 35 °N to 70 °N with a spatial resolution of 0.5 x 0.25 degrees lon-lat, roughly corresponding to 25 by 25 km. The model

has been applied in numerous studies for particulate matter (Schaap M. et al., 2004; Schaap M. et al., 2007). The modelled PM10 concentration over Europe with the LOTOS-EUROS model in the year 2003 is shown in figure 2. The modelled concentrations can be compared with measured concentrations and showed that the model follows the temporal patterns correctly but that the absolute concentrations are systematically underestimated. This is explained by the omission of natural sources other than sea salt and resuspension emissions in the model input. When a comparison is made for an almost exclusively anthropogenic component like PM2.5 sulphate aerosol, both absolute concentrations and temporal patterns are predicted correctly by the model.

The model input in this study are gridded maps of crustal material emitted as PM10 with 80 % of the particles in coarse mode (PM2.5 - 10) and 20 % of the particles belong to the fine mode (PM2.5). The tracers for crustal material in the model are chemically inert and deposition is modelled as fine and coarse mode particles. For a full model description we refer to the model documentation by Schaap M. et al. (2007).

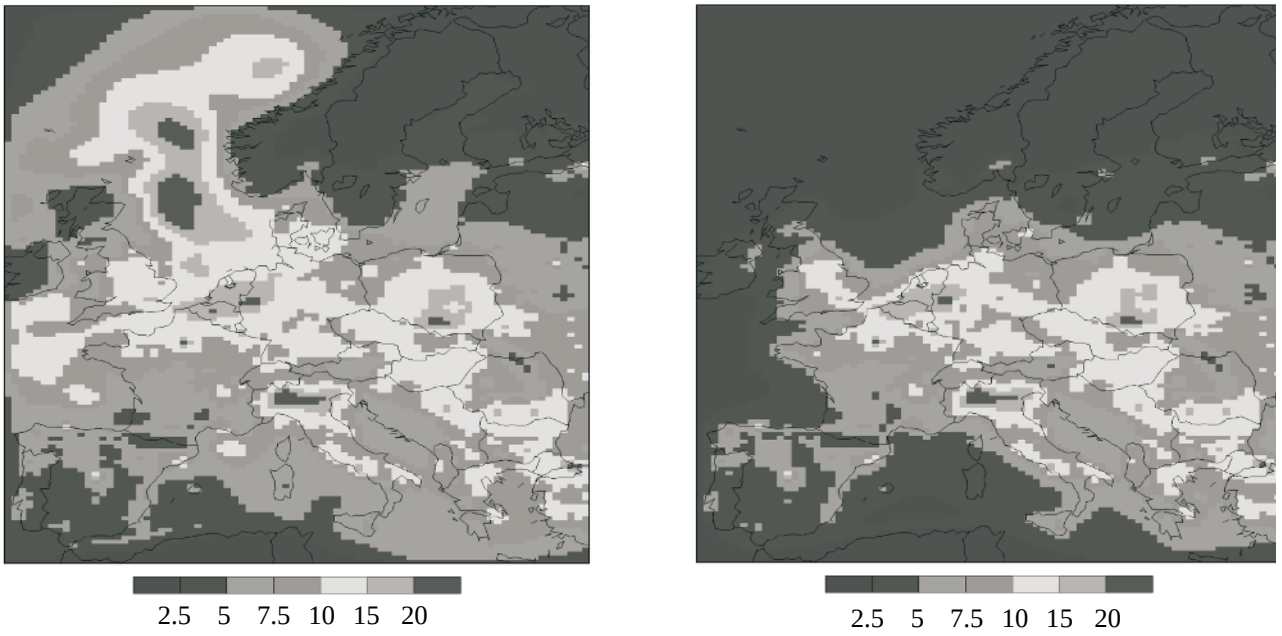

Figure 2:

Modelled PM10 concentration over Europe in 2003, Left PM10 including sea salt aerosol but excluding other natural soil dust emissions and resuspension; right anthropogenic PM10

5 Mining observational data to get additional information

In our approach (figure 1) we have too many unknowns to be directly successful. However we can derive important hints from the observational data. For example, a remarkable feature of the data compiled in table 1 is the strong elevation of crustal material at the kerbside stations, suggesting an important contribution of (resuspended) crustal material by traffic. This implies that if we intend to constrain e.g., the source strength of crustal material emissions due to wind erosion using models and observational data, this cannot be done without including traffic emissions.

5.1 Ambient PM10 concentration of crustal material in the Netherlands

For the Netherlands additional information about crustal material in ambient PM is derived from the data collected in the bronstof study (Visser H. et al., 2001). Visser H. et al. (2001) investigated the composition and origin of airborne particulate matter in the Netherlands by collecting samples of the fine fraction (PM2.5) and the coarse fraction (PM2.5 - 10) at six sites during one year (1998 - 99). The samples were analysed for chemical composition, for a description of the measurement procedures and analytical specifications we refer to Visser H. et al. (2001). These data will be used to gain information about the nature of

the CM sources contributing to ambient PM10 in the Netherlands. A number of elements are known to be present in crustal material, the most important being Si, Al, Ca, K, Fe and Ti The strong correlations between these elements in PM samples analyzed by Visser H. et al. (2001) suggest they originate from the same source, especially Al, Si and Ti (data not shown). Clay minerals in soils and rock have complex aluminosilicate structures and although measuring the aluminium and silicon content is relatively straightforward, the associated oxygen has to be estimated indirectly by assuming a certain oxidation state. Si and Al are the most abundant elements in crustal material and would be the most robust tracers. A formula [1] was derived, based on the chemical composition of the fine fraction of Dutch top soils to calculate the contribution of crustal material (CM) using the concentrations of Al and Si.

Mass CM = 0.49*[Si] + (2.36*[Si]+2.70*[Al]) [1]

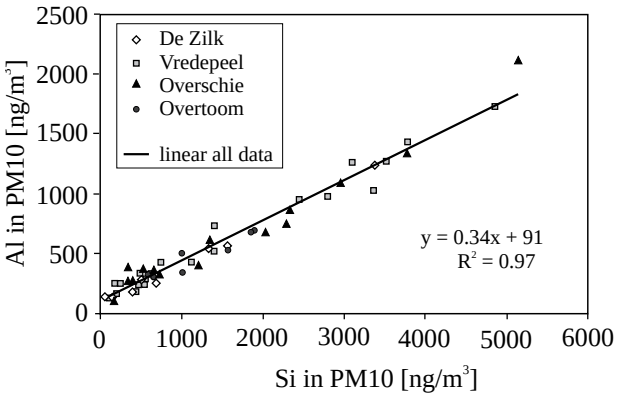

Figure 3:
Aluminum (Al) concentration as a function of Silicium (Si) in PM10 at four locations in the Netherlands (based on data from Visser H. et al., 2001)

The number of analysis days in the study of Visser H. et al. (2001) with a complete elemental analysis of PM is rather limited. To bypass the problem of limited data availability, the strong correlation between Al and Si in the available data (figure 3) is used to calculate soil material on the basis of Si alone if no Al data where available (table 2). The average concentration of CM in PM10 for the Netherlands is ~2.5 at the coast to ~ 5.5 µg/m³ at the inland sites.

The contribution of crustal material to ambient PM10 at the Dutch sites is wind direction dependent (data not shown). However, the number of observations available to calculate a wind direction specific contribution is limited. To approximate the wind direction dependency and at the same time cover a reasonable number of observations the samples have been split into a continental/land and sea derived origin (table 3). The air masses with a more continental/land origin show an elevated crustal material component compared to the sea derived air masses (2.9 vs 1.8 µg/m³ for de Zilk and 8.6 vs 3.0 µg/m³ for Vredepeel, respectively). This may help us to identify source regions using trajectories in the models for the particular days with elevated concentrations as well as investigating the dependency on meteorological conditions.

Table 2:
Average annual contribution of CM to ambient PM10 concentrations at four locations

Location method	DeZilk CM_alt	Vredepeel CM_alt	Overschie CM_alt	Overtoom CM_alt
number observations	31	45	42	49
avg (µg/m3)	2.4	5.4	5.0	3.3
sd	2.5	4.7	3.9	2.2

CM_alt = like CM eq[1] but missing Al data are estimated based on the Al to Si correlation (figure 3)

5.2 Size fractionation of crustal material in PM10

Another important feature that is needed for the models to predict the contribution of particulate emissions to ambient concentrations is the size fractionation. The smaller size fractions are transported of longer distances and have a longer life time due to a slower deposition rate. We have started our investigations assuming that 20 % of the CM is in the fine fraction (PM2.5) of PM10. These assumptions

Table 3:
The crustal material contribution to PM10 at de Zilk and Vredepeel at different wind directions

	De Zilk			Vredepeel		
	All	Continental	Sea	All	Continental	Sea
Wind direction	0 -360	(0 - 180)	(180 - 360)	0 - 360	(0 - 180, 315 - 360)	(180 - 315)
observations	31	18	13	45	19	26
Average (µg/m³)	2.4	2.9	1.8	5.4	8.6	3.0
stdev	2.5	3.1	1.4	4.7	4.8	2.9

can be refined using information from measurements. The average fraction of PM2.5 at the inland site (Vredepeel) and coastal site (De Zilk) is 15 % and 21 %, respectively. However, a closer look reveals that this fraction is not stable but changes as a function of the total CM content (figure 4). So, the more important the CM contribution, the less relevant is the PM2.5 fraction. This indicates that events of elevated CM contributions are probably highly controlled by sources in the vicinity. In future sensitivity studies we will vary the size fraction to see how this influences the sources contributing to our receptor points. We need to further analyze the data to see if we can use this information to exclude certain sources as being important for the Netherlands.

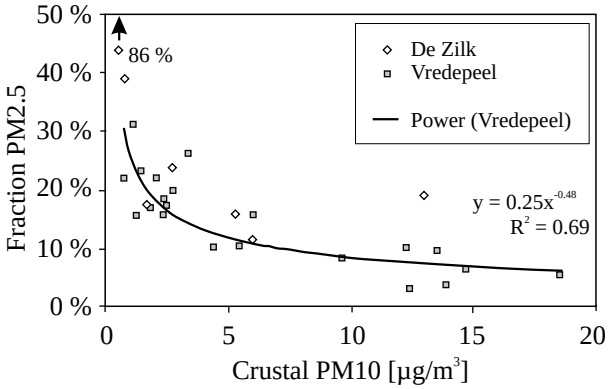

Figure 4:
The fraction of PM2.5 in crustal PM10 for two Dutch background stations

6 First results and further steps

The predicted concentration of crustal PM10 due to wind-blown dust from arable fields is presented in figure 5. This first approximation results in annual contributions to PM10 of ~ 0.2 µg/m^3 in the Netherlands. This is small compared to annual observed concentrations of 2 - 5 µg/m^3. However, it should be noted that our emission function does not yet include emission due to land management and, as was explained earlier, is highly uncertain. The patterns in the map reflect a combination of where arable lands are and arid regions because the emission is switched off on rainy days.

The predicted concentration of crustal PM10 due to re-suspension and roadwear by traffic is presented in figure 6. Here the patterns reflect where most kilometres are driven (big cities and densely populated regions) and predict a contribution of traffic to crustal PM10 in the Netherlands of 0.3 - 1 µg/m^3, again modest compared to the observed annual concentrations. Further investigations of published road side measurements may help us to better define realistic estimates of the resuspension emission factor and the fraction of crustal material therein. Furthermore we may vary the resuspension emission factor between highways, urban areas and rural, secondary roads. It is to be expected that a car or lorry driving over a rural road is causing more resuspending of dust than the same vehicle being one of the 4.000 vehicles / hr passing a busy highway. It should be investigated how sensitive the patterns of the predicted concentrations are to such modifications and how sensitive they are to certain climatic conditions.

7 Conclusions and Outlook

Crustal material is an important component of the PM10 and especially PM2.5 - 10 concentrations observed in the Netherlands. It originates from natural and anthropogenic sources. A better understanding of the sources contributing to the crustal fraction in PM10 is important for understanding the ambient PM concentrations because the fraction of PM that can be influenced by abatement measures in future policy plans should include the anthropogenic sources of the crustal fraction such as resuspension of dust by traffic.

Despite the importance of crustal material in total PM10 mass, the sources are still poorly understood and not well-represented in emission inventories. This is due to the fact that some sources can be defined as natural sources (e.g., erosion) whilst others like re-suspension are not recognized as primary emission but a re-emission. Separating the source contributions is difficult because the unique tracers for crustal material in PM10 samples (e.g., Si, Al, Ti) do not allow a distinction between the potential sources of this material. To make progress in our understanding of the crustal material source strengths we need to combine information from different disciplines. We present a methodology to combine the of information available from flux measurements, emission inventories, geographical information systems, atmospheric transport models,and chemical analysis of ambient PM10 samples to explore and constrain source strengths of the various sources of crustal PM10. Crustal material is dominantly found in the coarse fraction of PM10 (figure 4). Therefore, the coarse fraction (PM2.5 - 10) is the best fraction to study using the model. Even more so, because the coarse fraction is transported over shorter distances implying that the distance of the receptor site (where concentrations are measured) will be relatively close to source of origin.

Source functions and emission maps for other crustal PM10 sources need to be made and the predicted concentrations can be superimposed on each other. Given the uncertainty is our first approximations and that the sources will add up we can conclude that it is certainly feasible to come close to observed values. For most of the crustal

PM10 sources climatic conditions like precipitation and wind are controlling factors. The atmospheric dispersion models can be run with different years to match the moments of campaigns with sufficient chemical analytical data. For example, the maps in figure 5 and figure 6 are the result of using the meteorological data of 1998 and 1999 to match the sample dates of the Visser et al. (2001). In this way we may be able to use chemical composition data for different years from different regions.

The temporal and spatial patterns of observations and model predictions will give important clues to whether the balance between sources and the timing of there emissions is correct in our appraoch. Within the uncertainties surrounding this exploratory approach the balance in source contributions can be optimized to fit the observations in terms of absolute annual concentrations as well as the temporal patterns over the year. The result is a constraint on the total source strength of crustal material and an indicative ranking of the importance of contributing sources in various areas of Europe.

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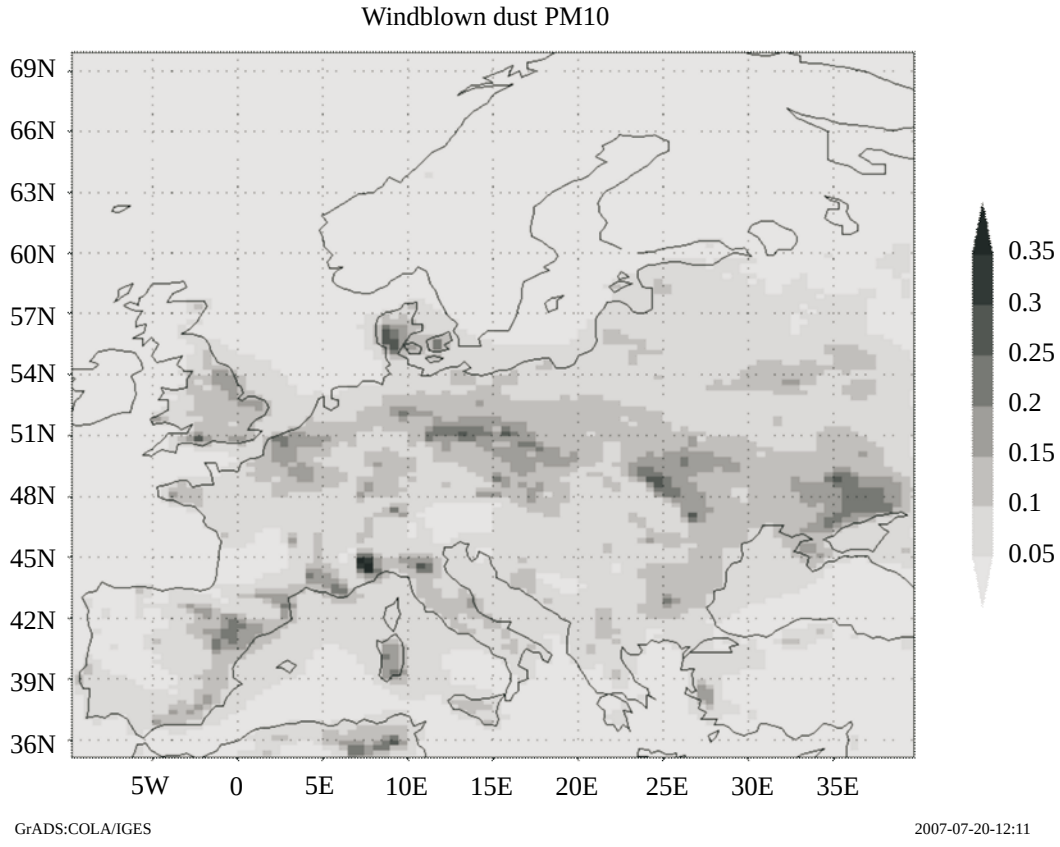

Figure 5:
Predicted crustal PM10 over Europe using a first approximation of the windblown dust from arable lands

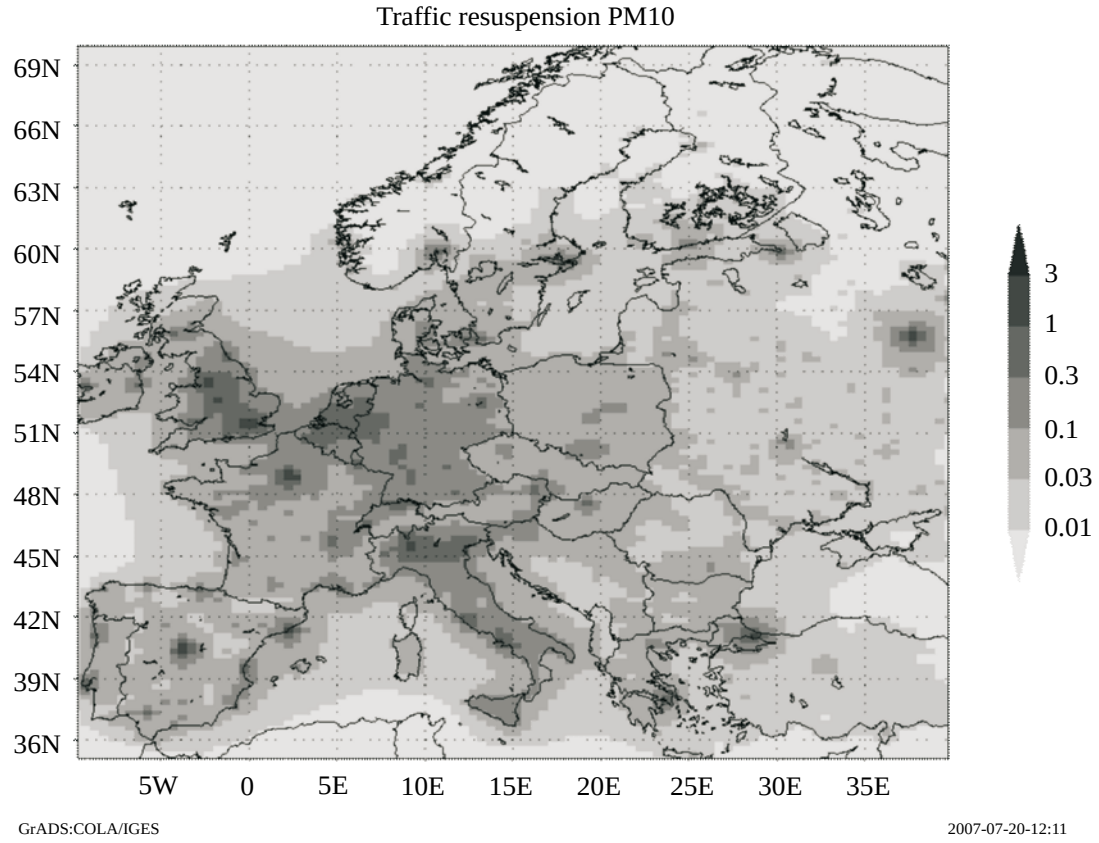

Figure 6

Predicted crustal PM10 over Europe using a first approximation of resuspended crustal material by traffic

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PM emission factors for farming activities by means of dispersion modeling

D. Öttl[1] and R. Funk[2]

Abstract

In this study emission factors for use in emission inventories are provided for different agricultural operations. These are plowing, harrowing, disking, and cultivating. Emission factors were derived for PM10, PM2.5, and PM1. Measurements were conducted by the Leibniz-Centre for Agricultural Landscape Research (ZALF) in Müncheberg, Germany, while the emission factors were obtained with the Lagrangian dispersion model GRAL (Graz Lagrangian Model). The latter has been developed by the Institute for Internal Combustion Engines and Thermodynamics, Graz University of Technology, Austria.

Keywords: agricultural operations, PM emission factors, Particle model, farming activities

Introduction

Fine particulate matter (PM) is nowadays being recognized as one of the most critical pollutants regarding the compliance with air quality standards. In many studies it became evident, that the major source for high PM-concentrations close to the ground is traffic. However, it is recognized that there are many other sources (anthropogenic and natural), which can also contribute significantly to the total pollutant burden. Air quality observations indicate that the so called background concentration of PM10 accounts for roughly 50 % of the total observed concentrations even within larger cities (Lenschow P. et al. 2001). In order to allow for a detailed source apportionment regarding the background concentration, comprehensive emission inventories are necessary. There exist only a few studies about PM emission factors from agricultural operations (Holmén B.A. et al. 2001), and hence, these are not accounted for in emission inventories. This study aims at providing emission factors for different kinds of agricultural operations namely: plowing, harrowing, disking, and cultivating.

Experimental set up

The experiments were all conducted by the Leibniz-Centre for Agricultural Landscape Research (ZALF) in Müncheberg, Germany. The observations consisted of

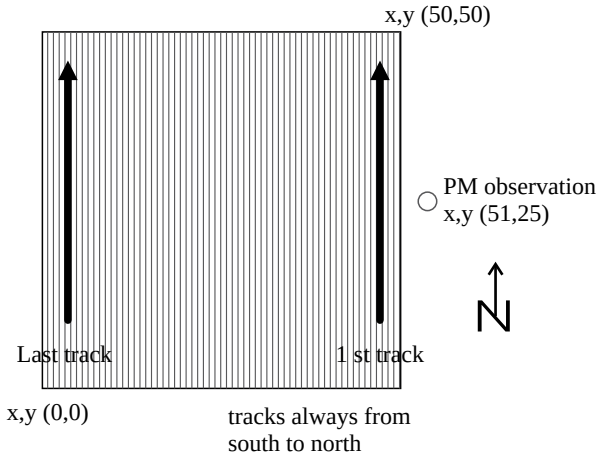

Figure 1:
Sketch of the test field for the different agricultural operations

[1] Air Quality Department of Styria, Austria

[2] Leibniz-Centre for Agricultural Landscape Research (ZALF) Müncheberg, Germany

a wind vane and a cup anemometer 1 m above ground level, and a particle counter for PM10, PM2.5, and PM1 (GRIMM 107 PTFE ENVIRONcheck). PM observations took place at a height of 2 m above ground level close to a test field, which had an extension of 50 m x 50 m (figure 1). In all experiments the tractor drove from south to north.PM emission factors for farming activities by means of dispersion modeling

Figure 2 shows the investigated agricultural operations. The soil type of test field can be characterized as sandy cambisol. The soil texture is given in table 1. It can be seen, that PM10 accounts for about 6 % of the total volume. Figure 3 depicts a typical soil moisture distribution within the test field during the experiments. Little variation was found for the experiments, which took place between June and October 2004. It is clearly visible, that only the first few centimeters of the soil may significantly contribute to PM emissions, as the soil moisture is strongly increasing with depth up to a constant value of about 5.5 %. Tests in a wind tunnel revealed a critical value of about 2.5 % soil moisture above which practically no PM emission takes place.

Table 1:
Soil texture of the test field

Fraction	Diameter (µm)	Share (%)
Coarse sand	630 - 2000	5.0
Middle sand	200 - 600	37.9
Fine sand	63 - 200	40.4
Coarse silt	20 - 63	7.5
Middle silt	6 - 20	3.3
Fine silt	2 - 6	1.8
Clay	<2	4.2

Figure 2:
Investigated agricultural operations: plowing including a packer, harrowing, disking, and cultivating

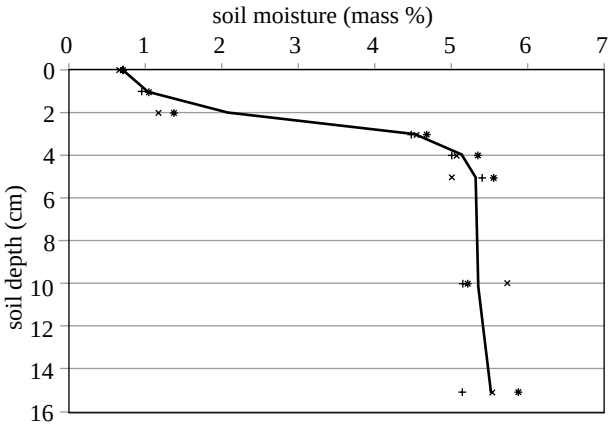

Figure 3:
Typical soil moisture distribution within the test field during the experiments

Modeling approach

In order to obtain PM emission factors the Lagrangian dispersion model GRAL (Graz Lagrangian Model) has been utilized. GRAL is a well-validated short range numerical model applicable for a wide range of wind speeds and atmospheric stabilities. In this study the following relationships were used to determine the turbulent velocities:

$$du = -(pu + qv)dt + \sigma_u \sqrt{2pdt}\,\xi_u \tag{1a}$$

$$dv = -(-qu + pv)dt + \sigma_v \sqrt{2pdt}\,\xi_v \tag{1b}$$

$$dw = a \cdot dt + \left[C_0 \cdot \varepsilon\ dt\right]^{0.5} \xi_w \tag{1c}$$

du, *dv*, and *dw* are the wind fluctuations in x, y, and z-direction. ξ_u, ξ_v, and ξ_w are increments of a Wiener process with zero mean, a standard deviation of one and a Gaussian probability density function. *dt* is the time step, ε the ensemble average rate of dissipation of turbulent kinetic energy, C_0 the universal constant set equal to 4 (Wilson J.D. et al. 1996, Degrazia G. A. et al. 1998, Anfossi D. et al. 2000), a is the deterministic acceleration term computed according to Franzese (Franzese P. et al. 1999), and $\sigma_{u,v}$.are the standard deviations of the horizontal wind fluctuations. The latter are determined in the surface layer by

$$\sigma_{u,v} = \mathrm{u}_* \cdot \left(1 + 1.8 \cdot \left|\frac{\mathrm{z}}{\mathrm{L}}\right|^{0.1}\right) \tag{2}$$

The ensemble average rate of dissipation of turbulent kinetic energy according to Kaimal and Finnigan (Kaimal J. C. et al. 1994) is:

$$\varepsilon = \frac{\mathrm{u}_*}{\kappa z}\left(1 + 0.5\left|\frac{z}{L}\right|^{0.67}\right)^{1.5} \tag{3}$$

In eq. (2) and (3) u_* denotes the friction velocity, κ the von Karman constant, and *L* is the Monin-Obukhov length. The friction velocity is determined according to Golder (Golder D. 1972) and the Monin-Obukhov length according to Venkatram (Venkatram A. 1996).

Equations (1a) and (1b) were derived by Oettl (Oettl D., et al. 2005) to account for horizontal meandering flows in low wind speed conditions. The parameters *p* and *q* control the shape of the modelled autocorrelation function, which usually shows oscillation behaviour in low wind speed conditions and an exponential shape in higher wind speed conditions (Anfossi D. et al. 2004, Hanna S. R. 1983). According to Anfossi (Anfossi D. et al. 2004) parameters *p* and *q* are defined as:

$$p = \frac{2}{\left(m^2 + 1\right) \cdot T_3} \tag{4a}$$

$$q = \frac{m}{\left(m^2 + 1\right) \cdot T_3} \tag{4b}$$

$$T_3 = \frac{T_* \cdot m}{\left(m^2 + 1\right) \cdot 2\pi} \text{ for mean wind speeds } \bar{u} < 2.5 \text{ m s}^{-1} \text{ and} \tag{4c}$$

$$T_3 = \frac{T_* \cdot m}{\left(m^2 + 1\right) \cdot 2\pi} \text{ for } \bar{u} \geq 2.5 \text{ m s}^{-1} \tag{4d}$$

$$T_* = 200 \cdot m + 350 \tag{4e}$$

$$m = \frac{8.5}{\left(\bar{u} + 1\right)^2} \text{for } \bar{u} < 2.5 \text{ m s}^{-1}, \text{ and m = 0 for } \bar{u} \geq 2.5 \text{ m s}^{-1} \tag{4f}$$

The empirical relationships for T_* and m are based on observations using a sonic anemometer in the city of Graz (Austria). For higher wind speeds ($\bar{u} \geq 2.5$ m s^{-1}) eq. (1a and 1b) collapse on the classical Langevin equation for homogeneous turbulence.

A direct assessment of the uncertainties related with the dispersion model is impossible. In order to obtain some estimate a comparison of modeled concentrations with GRAL and observations during the Prairie Grass experiment was made. The Prairie Grass experiment (Barad M. L. 1958) included 10-minute near-surface sampling along five arcs, 50 to 800 m, downwind from a near-surface point source release of sulfur dioxide. The 20-minute releases were conducted during July and August 1956, with an equal number

of cases occurring during the daytime and nighttime. The sampling was for the 10-minute period in the middle of the 20-minute release. All in all 44 experiments were used in this study. As our interest is on the model performance very close to the source, only the results for the 50 m sampling arc are briefly discussed. Figure 4 depicts a comparison of observed and modeled maximum concentrations at 50 m distance from the release point. The lines indicate the one to one relationship and a deviation of +/- 30 %. As can be seen almost all cases fall within this range of uncertainty. The coefficient of determination was found to be 0.94.

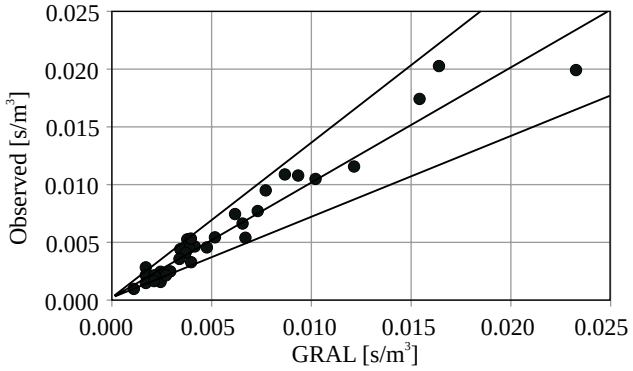

Figure 4.

Observed and modeled maximum concentrations at 50 m distance from the release point for the Prairie Grass Experiment (44 cases)

Results

Emission factors for PM were obtained by modelling the dispersion from the test field, treating it as an area source. It was assumed in the simulations, that the PM emissions are initially mixed up to 2 m above ground level due the tractor induced turbulence. Figure 5 shows an example of observed PM10-concentrations for plowing and packing. From the observed concentration minima during the experiment, the background concentrations for PM10, PM2.5, and PM1 were determined.

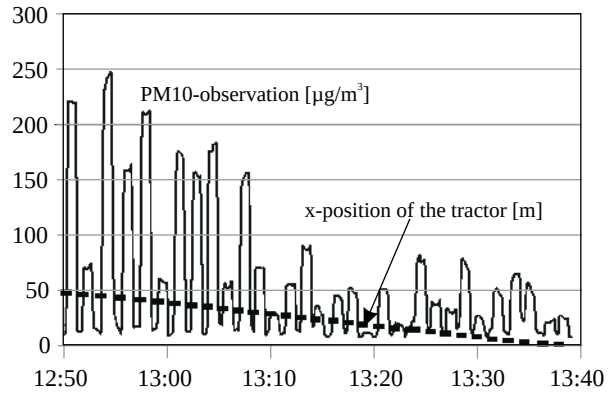

Figure 5:

Example of observed PM10-concentrations for plowing and packing

For each experiment an hourly average concentration was calculated by integrating the observed peaks and subtracting the estimated background concentration. From the dispersion simulation, emission factors in units of [kg h^{-1}] and [kg] were obtained respectively. By division through the area of the test field the final emission factors in units of [mg m^{-2}] could be derived. Table 2 lists the observed meteorological conditions during the tests. Although the stability class is subject to some uncertainty as it had to be estimated based on the wind speed, cloud cover, time of the day, and season, model simulations applying other stability classes showed only little influence on the concentration. The reason is the very close location of the PM measurement site to the test field.

Table 2:

Observed meteorological conditions during the experiments

	Wind-speed [m s^{-1}]	Wind direction [deg.]	Stability class (PGT)	Relative humidity [%]	Temperature [°C]
Plowing and packing	2.7	170	C	40	24
Harrowing	1.9	212	B	34	27
Disking	3.1	96	D	45	13
Cultivating	1.1	113	B	29	27
Plowing	2.9	13	D	53	27

Table 3 lists the calculated emission factors for the different agricultural operations for PM10, PM2.5, and PM1. Having in mind all the possible influencing factors on the emissions such as dispersion model uncertainty, wind speed, relative humidity, soil moisture, land preparation, it is somewhat surprising to find with one exception a relatively narrow range for the emission factors. However, during dry conditions emissions for similar activities can rise up by a factor of 10 as can be seen for plowing.

Table 3:

Emission factors for PM10, PM2.5, and PM1 for different agricultural operations

	PM10 [mg m^{-2}]	PM2.5 [mg m^{-2}]	PM1 [mg m^{-2}]
Plowing and packing	120	5	1
Harrowing	82	29[1)]	<1
Disking	137	12	3
Cultivating	186	6	2
Plowing (dry conditions)	1045	129	13

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List of speakers

Name	Title	Institution	Adress Telephone Fax	E-mail
Aneja, Viney P.	Prof. Dr.	Department of Marine, Earth, and Atmospheric Sciences	Room 5136, Jordan Hall Campus Box 8208 North Carolina State University Raleigh NC 27695-8208, U.S.A. +01 (919) 515-7808 +01 (919) 515-7802	VINEY_ANEJA@NCSU.edu
Beletskaya, Olga		University Hohenheim Institute of Farm Management (410B)	Scharnhauser Straße 79 D 73760 Ostfildern, Germany +49 711 459 22 567	kate_mure@hotmail.com
Bünger, Jürgen	Dr.	Berufsgenossenschaftliches Forschungsinstitut für Arbeitsmedizin (BGFA) Institut der Ruhr-Universität Bochum	Bürkle-de-la-Camp-Platz 1 D 44789 Bochum, Germany +49 (0)234 302-4556 +49 (0)234 302-4505	buenger@bgfa.de
Denier van der Gon, Hugo A. C.	Dr.	TNO Built Environment and Geosciences Business unit Environment and Health	Iaan van Westenenk 501 NL-7300 AH Apeldoorn The Netherlands +31 55 5493 267 +31 55 5493 252	Hugo.deniervandergon@tno.nl
Funk, Roger	Dr.	ZALF Müncheberg Institut für Bodenlandschaftsforschung	Eberswalder Straße 84 D 15374 Müncheberg, Germany +49 33432 82321	rfunk@zalf.de
Grimm, Ewald		Kuratorium für Technik und Bauwesen in der Landwirtschaft e. V. (KTBL)	Bartningstraße 49 D 64289 Darmstadt, Germany +49 6151 7001-156 +49 6151 7001-123	e.grimm@ktbl.de
Gustafsson, Gösta	Prof.	Dept. of Agricultural Biosystems and Technology	P.O. Box 86 S 23053 Alnarp, Schweden +46 404 15488 +46 404 154 75	gosta.gustafsson@jbt.slu.se
Hartung, Jörg	Prof. Dr.	Institut für Tierhygiene, Tierschutz und Nutztierethologie Tierärztliche Hochschule Hannover	Bünteweg 17p D 30559 Hannover, Germany +49 511 953 8832 +49 511 953 8588	itt@tiho-hannover.de
Haunold, Werner		Institut für Atmosphäre und Umwelt Universität Frankfurt	Altenhöferallee 1 D 60438 Frankfurt, Germany +49 69 798 40 239	haunold@iav.uni-frankfurt.de
Hinz, Torsten	Dr.	Institut für Technologie und Biosystemtechnik Bundesforschungsanstalt für Landwirtschaft (FAL)	Bundesallee 50 D 38116 Braunschweig Germany +49 531 596 4202 +49 531 596 4299	torsten.hinz@fal.de
Ruschel, Yvonne				yvonne.ruschel@fal.de
Hnilicova, Helena		Czech Hydrometeorological Institute	Na Sabatce 17, 143 06 Praha 4, Czech Republic +420 244 032 419 +420 244 032 468	Hnilicova@chmi.cz
Keder, Josef	Dr.			keder@chmi.cz
van der Hoek, Klaas W.		National Institute for Public Health and the Environment (RIVM)	P. O. Box1 NL 3720 BA Biltkoven The Netherlands	Klaas.van.der.Hoek@rivm.nl
Karlowski, Jerzy		Institute for Buildings Mechanisation and Electrification of Agriculture	ul. Biskupinska 67 PL 60-463 Poznan, Poland	j_karlowski@poczta.onet.pl , jkarlo@ibmer.waw.pl
Klimont, Zbigniew		International Institute for Applied Systems Analysis	Schlossplatz 1 A-2361 Laxenburg, Österreich +43 2236 807 547 +43 2236 807 533	klimont@iiasa.ac.at
Lindenthal, Gerfried	Dr.	Palas GmbG	Greschbachstraße 3b D 76229 Karlsruhe, Germany +49 721 96213 0 +49 721 96213 33	mail@palas.de
Schmidt, Martin				
Nannen, Christoph		Institut für Landtechnik, Universität Bonn	Nußallee 5 D 53115 Bonn, Germany	c.nannen@uni-bonn.de
Rosenthal, Eberhard				rosenthal@uni-bonn.de
Madsen, Peter Vangsbo	Project-management	National Environmental Research Institute (NERI)	Frederiksborgvej 399, 4000 Roskilde, Denmark +45 4630 1154 +45 4530 1214	pvm@dmu.dk
Öttl, Dietmar	Dr.	Amt d. Steiermärkischen Landesregierung FA17C Referat für Luftgüteüberwachung	Landhausgasse 7 A 8010 Graz, Österreich +43 316 877 3995	dietmar.oettl@stmk.gv.at
Romann, Martin		Sympatec GmbH	Am Pulverhaus 1 D 38678 Clausthal-Zellerfeld Germany +49 5323 717-234 +49 5323 717-229	mromann@sympatec.com
Schärer, Bernd		Umweltbundesamt - Federal Environment Agency	Postfach 1406 D 06813 Dessau, Germany +49 340 2103 2368 +49 340 2104 2368	bernd.schaerer@uba.de
Schneider, Freidhelm	Dr.	Grimm Aerosol Technik GmbH & Co. KG	Dorfstraße 9 D 83404 Ainring, Germany +49 8654 578 22 +49 8654 578 35	fsn@grimm-aerosol.com
Takai, Hisamitsu		Department of Agricultural Engineering Research Centre Bygholm	Schüttesvej 17 DK 8700 Horsens, Danmark	Hisamitsu.Takai@agrsci.dk

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