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Atmospheric nitrogen dynamics in Hesse, Germany: The challenge and its potential solution

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Abstract

Reactive atmospheric nitrogen species contribute to adverse effects in ecosystems (acidification and eutrophication) and in the troposphere (ground level ozone and secondary particle formation, both leading to further adverse effects to ecosystems and human health). However, dose-response relationships are not at all well established. In a comprehensive approach for the German federal state of Hesse, emissions, atmospheric concentrations and chemistry as well as deposition fluxes are assessed by a matching set of models and measurements, aiming at a regionally closed nitrogen balance. Measurements cover all reactive species, the respective source and sink properties and the meteorological parameters relevant for model operation and validation: gaseous ammonia, nitric oxide, nitrogen dioxide, nitrous and nitric acids as well as ammonium and nitrate in aerosols and sedimenting particles. This paper describes the challenge, i.e. the present deficits in knowledge, and derives a methodological approach and the practical means applied or planned (locations, equipment, procedures) in principle.

Key words: nitrogen balance, emissions, atmospheric transport, depositions, experimental design, modelling

Atmosphärische Dynamik von Stickstoff in Hessen - Herausforderung und Lösungsmöglichkeiten

Zusammenfassung

Reaktive Stickstoff-Verbindungen tragen zu unerwünschten Wirkungen in Ökosystemen (Versauerung und Eutrophierung) und in der Troposphäre bei (bodennahes Ozon und Bildung von Sekundär-Aerosolen; beide beeinträchtigen Struktur und Funktion von Ökosystemen und die menschliche Gesundheit). Im Vergleich zur Bedeutung der Probleme ist über Dosis-Wirkung-Beziehungen wenig bekannt. In einem umfassenden Ansatz soll für das Gebiet des Bundeslandes Hessen versucht werden, Emissionen, Konzentrationen und Chemie in der bodennahen Atmosphäre und Depositionen im Hinblick auf eine geschlossene Stickstoff-Bilanz mit Hilfe von aufeinander abgestimmten Modellen und Messungen zu quantifizieren. Die Messungen umfassen alle reaktiven Spezies, ihre Quellen und Senken sowie die meteorologischen Parameter, die zum Betrieb der Modelle und zu ihrer Validierung erforderlich sind. Sie umfassen die Gase Ammoniak, Stickstoffmonoxid, Stickstoffdioxid, salpetrige Säure und Salpetersäure sowie Ammonium und Nitrat in Aerosolen und sedimentierenden Partikeln. Dieser Beitrag beschreibt den "Stand des Unwissens" als Herausforderung, leitet daraus einen methodischen Ansatz sowie die praktischen Maßnahmen zur Umsetzung ab.

Schlüsselworte: Stickstoff-Bilanz, Emissionen, atmosphärischer Transport, Depositionen, experimentelles Konzept, Modellierung

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1 The challenge

Although classical air pollution problems caused by very high concentrations of sulphur dioxide and London type smog seem to have been decreased to acceptable levels, adverse effects such as acidification and eutrophication of natural and semi-natural ecosystems, increased tropospheric ozone concentrations and stratospheric ozone depletion as well as greenhouse effects and human health problems caused by aerosols are still potential threats. Atmospheric nitrogen species contribute to all phenomena (Dämmgen and Sutton, 2001; Erisman et al., 1998b; Graedel and Crutzen, 1995).

Reactive nitrogen can be formed by combustion processes, by industrial ammonia production or biological N-fixation. Regionally, another source of reactive nitrogen can be the import of nutrients through feed used for intensive livestock breeding. Organic nitrogen compounds (manure) originate from both anthropogenic sources as well as natural sources.

As illustrated in Figure 1, about half of the potential acidifying input in a north German forest (Schorfheide) can be attributed to nitrogen species; inputs of oxidized nitrogen species (gas: nitric acid - HNO_3 ; aerosol constituent and bulk deposition: nitrate - NO_3^-) fall below those of reduced nitrogen (gas: ammonia - NH_3 ; aerosol constituent and bulk deposition: ammonium - NH_4^+). Obviously, the acidity derived from reduced nitrogen deposition exceeds the critical level for the pine forest ecosystem considered. In Germany, acidity inputs are beyond critical loads for 90 % of the forested area (Nagel & Gregor, 1999).

Also, atmospheric N inputs fertilize natural and semi-natural ecosystems to an extent that their natural structure and function are strained (Table 1): eutrophication abatement is thought to be the more severe future problem, because sulphur dioxide emissions will continue to decrease.

Most of the aerosol matter is in the $\text{PM}_{2.5}$ fraction, which is almost entirely composed of NH_4^+ salts formed by chemical reaction of NH_3 and atmospheric acids (Zinder et al., 1988; ten Brink et al., 1996; Dämmgen, 2002; Harrison et al., 2002; Hass et al., 2002; Spindler et al., 2002). They are so-called secondary particles. From Table 1 one can deduce that approx. half of the aerosol- NH_4^+ is NH_4NO_3 . NH_4Cl is stable at temperatures below 10 °C (e.g. Saitoh et al., 2002), only. In Central Europe and the Mediterranean area its contribution to aerosol- NH_4^+ is almost negligible. This means that half of the aerosol NH_4^+ is found as ammonium sulfates. As shown in Figure 2, aerosol-S concentrations have not diminished during the past decades despite of considerable reductions of SO_2 emissions; secondary aerosol formation is governed by the availability of atmospheric NH_3 and thus by NH_3 emissions. Therefore, the problem of air pollution abatement with regard to aerosols is strongly connected with NH_3 emission abatement.

In the polluted atmospheres of Central Europe, nitrogen oxides play a key role in the formation and destruction of ground level O_3 (Dämmgen et al., 1994a; Fowler, 1997). On wet surfaces, such as soil surfaces, nitric oxide (NO) is at least partly transformed to nitrous acid (HONO) which contributes to the formation of OH-radicals, especially in the morning (Zimmerling and Dämmgen, 2000).

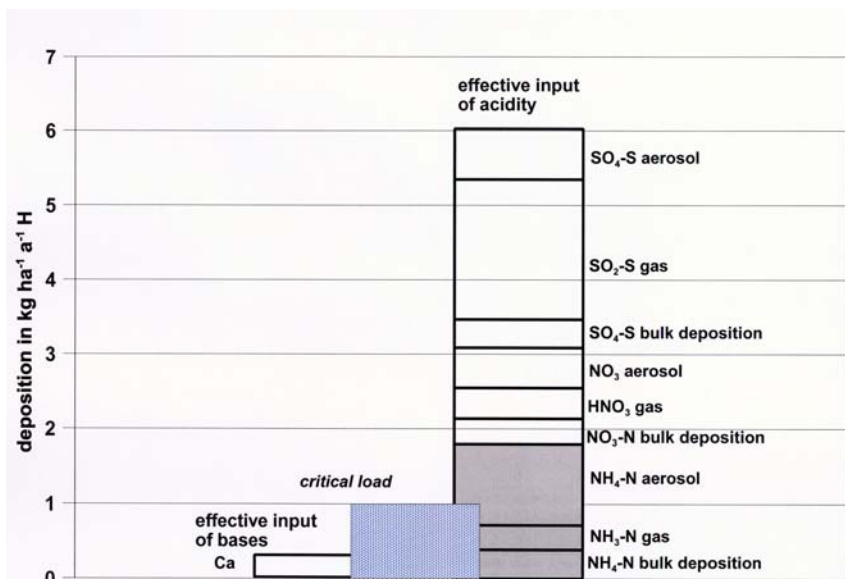


Fig. 1 Input of acidity into a pine forest ecosystem (Schorfheide, Brandenburg, Germany) due to deposition of reactive nitrogen and sulphur species (Dämmgen and Zimmerling, 2002b). The critical load for acidification was derived from Nagel and Gregor (1999)

Table 1

Inputs of reactive nitrogen species into a pine forest ecosystems (Speuld, The Netherlands - Erisman et al., 2001; Schorfheide, Brandenburg, Germany; Dämmgen and Zimmerling, 2002b), grassland ecosystems (Merenschwand, Switzerland - Hesterberg et al., 1996; Melpitz, Germany - Erisman et al., 2001) and a moorland (Auchencorth, Scotland - Erisman et al., 2001). Critical loads are derived from Nagel and Gregor (1999) and Hesterberg et al. (1996)

	Speuld NL forest	Britz D forest	Merenschwand CH Grassland kg ha ⁻¹ a ⁻¹ N	Melpitz D grassland	Auchencorth GB moorland
NH ₃	20.8	6.8	12.9	7.3	2.5
NH ₄ ⁺ - aerosol	4.7	15.2	1.0	0.7	0.3
NH ₄ ⁺ - bulk	11.3	5.4	10.6	4.7	2.4
NO _x	2.8	1.0	3.4	0.2	0.6
HNO ₃ + HNO ₂	6.1	7.4	1.4	1.3	0.5
NO ₃ ⁻ aerosol	3.7	7.5	-	0.3	0.2
NO ₃ ⁻ bulk	5.3	4.8	3.4	3.3	1.9
Total	54.7	48.1	32.7	17.8	8.4
Critical load	-	5 - 10	15	-	-

Due to the wide range of sources of the various nitrogen species, their complicated atmospheric chemistry, the resulting short- and long-range transports and bi-directional vertical fluxes, atmospheric balances of reactive nitrogen are far from being understood in contrast to other atmospheric compounds which are only deposited, and can therefore be described more easily.

At present, the critical loads discussed and applied are more or less empirical (UBA, 1996). Critical load exceedances are modelled using modelled concentration fields which rely on modelled atmospheric chemistry and transport processes based on modelled emissions. Comparisons between modelled and measured doses (depositions) demonstrate that the present knowledge is unsatisfactory, in particular for reduced nitrogen species (Bleeker et al. 2000, Erisman and Draaijers, 1995; Jakobsen et al., 1996; Vonk et al., 2000). According to Erisman et al. (1998a) and Sutton et al. (2002) the major source of uncertainty in quantification of regional deposition esti-

mates is the lack of knowledge on the “exact” quantities of the respective emissions and their spatial and temporal variation. Even for the air pollutant known best, sulphur dioxide, emissions and depositions fall apart by the factor of two (Erisman and Draaijers, 1995; Jakobsen et al., 1996), for cadmium by the factor of four (Dämmgen et al., 2000). For sulphur dioxide, this is mainly due to the simplification of the deposition process in models and the small scale effects in concentrations and deposition, which are not incorporated in models. For cadmium, emission inventories seem to be inadequate (Ilyin et al., 2000).

Annual average concentrations of gaseous pollutants which are not influenced by local sources can be modelled with reasonable accuracy on a large scale. Therefore, country average emissions and deposition are accurate enough for policy negotiations, such as under the UN/ECE Convention on Long-Range Transboundary Air Pollution (CLRTAP) (UNECE, 1999; e.g. Vestreng and Støren, 2000). Locally, at ecosystem level, the uncertainty is much higher (Erisman and Draaijers, 1995).

Measured data suitable to validate the models, i.e. atmospheric concentration data for aerosol constituents, ammonia and nitric acid, are lacking in most of Central Europe, in particular for Germany (Schaap et al., 2002).

This paper provides the outlines of a comprehensive approach towards a stepwise solution of a quantification of the fluxes and pools of reactive nitrogen species in the atmosphere near the ground for a region of 21,114 km² in the low mountain range in Central Europe.

2 A stepwise approach to the solution

In Germany it is the task of the single federal states (Bundesländer) to practically assert air pollution control measures and to survey the results of these measures with

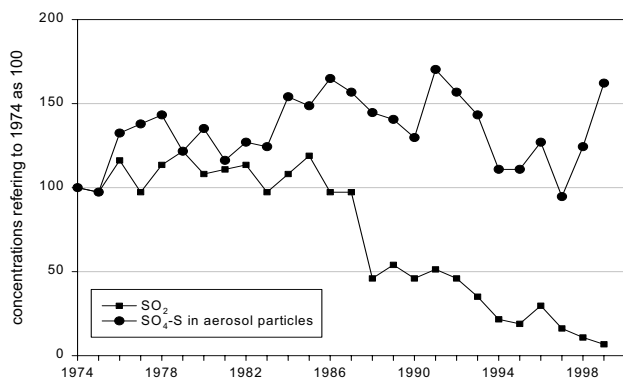


Fig. 2 Trends in the concentrations of SO₂ and aerosol-S in Germany (UBA, 2001)

adequate means, i.e. by measuring atmospheric concentrations and depositions. In Germany, adequate concentration measurements exist for sulphur dioxide (SO₂), the nitrogen oxides NO and nitrogen dioxide (NO₂) and ozone (O₃). Bulk and wet-only depositions lack accuracy in particular for reduced nitrogen (Draaijers et al., 2001; Dämmgen et al., 2003). Although, dry deposition monitoring is possible in principle (Hensen et al. 1999, Erisman et al., 2001), it is not undertaken by the official monitoring networks of the federal states.

The Hessian Agency for Environment and Geology have recognized that all attempts to solve these problems will remain incomplete unless a comprehensive and detailed approach can be realized. Therefore, within the Hessian programme "Digitale Umweltanalyse Hessen" (Hessian Digital Environmental Analysis, DUAH) the project "Atmospheric Nitrogen Dynamics and Budgets" was launched, which is to deliver a scientific base for political and administrative decisions. In particular, it is to identify and evaluate optimal abatement measures and the resulting conflict potentials.

Figure 3 summarizes the various relations between elements of such a comprehensive approach deducing a cause-effect relationship using both measurements and models: concentrations of gases and aerosols as well as relevant meteorological parameters measured locally are combined to form concentration fields taking into account atmospheric chemistry and transport phenomena. The sink properties of terrestrial and aquatic systems are primarily characterised by land use. Together with the measured deposition of sedimenting particles (bulk and wet-only

deposition), regional dry deposition of gases and aerosols can be modelled. Emission inventories with a high resolution in time and space for all relevant source categories yield emission budgets which are then compared with the deposition budgets. Transboundary fluxes can be identified. Risks are assessed by comparison of local deposition patterns and the properties of the aquatic and terrestrial sink system, in particular their critical loads. Abatement potentials can be identified which have to be discussed with regard to their ecological, economical and social benefits and potential conflicts.

2.1 Creating an emission data base

According to Directive 2001/81/EC, emission inventories serving the needs of the European legislation have to use the latest edition of the EMEP/CORINAIR emission inventory guidebook (Richardson, 2000). This guidebook includes the respective IPCC calculation procedures. Wherever appropriate, national data and detailed methodologies have to be used within the framework of the EMEP/CORINAIR instructions. In this case they have to be well documented.

Also, inventories have to be provided for national purposes following the principles of Bundes-Immissionsschutzgesetz (BImSchG, 1990), the relevant decrees (Verordnung zur Durchführung des Bundes-Immissionsschutzgesetzes, BImSchV) and administrative directives (Verwaltungsvorschrift zum Bundes-Immissionsschutzgesetz, BImSchVwV).

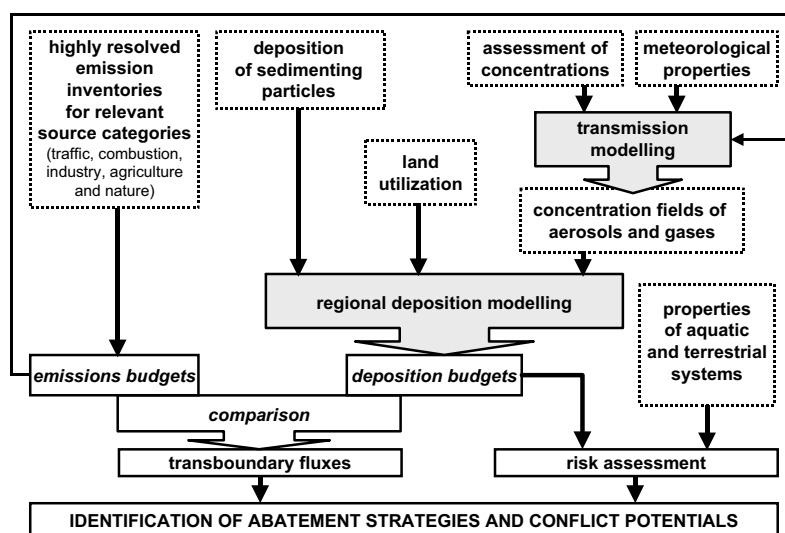


Fig. 3 Atmospheric nitrogen dynamics and budgets and their interrelations and assessment (squares with dotted margins: measured or modelled input data; grey arrows: atmospheric chemistry, dispersion, transport and regional deposition models; squares and arrow with words in italics: budgets and balances; square with words in capital letters: recommendations)

For the emission of nitrogen species, German inventories so far differentiate between industrial sources requiring official approval (genehmigungsbedürftige Anlagen, Industrie), minor commercial enterprises not requiring official approval (nicht genehmigungsbedürftige Anlagen, Kleingewerbe), domestic heating not requiring official approval (nicht genehmigungsbedürftige Feuerungen, Gebäudeheizung), traffic including automobiles, ships and planes (Verkehr) and biogenic and diffuse sources including agriculture, nature and waste management (biogene und nicht gefasste Quellen).

The emission inventory has a resolution in space of 1 km², single local communities (Städte, Gemeinden) or districts (Landkreise) depending on the respective source category. For nitrous oxide (N₂O) no resolution in space nor in time is necessary as N₂O is chemically inert in the troposphere. Its only known sink of importance is the stratosphere. For NO_x and NH₃ atmospheric lifetime is so short that an adequate treatment of their atmospheric dynamics presupposes a high resolution in space and time. In general, both seasonally/monthly and diurnal variation have to be taken into account. As illustrated in Figure 4, approx. 40-50 % of NH₃ emitted are deposited within the first 10 km. In order to connect emission, deposition and potential effects, the resolution in space must be in the same order of magnitude. For Germany this means, at least a resolution of single local communities.

At present, the Hessian emission inventory for all sources apart from biogenic emissions makes use of a more or less basic methodology combining fixed emission factors for a comparatively small number of categories with activity data according to HLUg handbook (in preparation). For biogenic emissions, the EMEP/CORINAIR guidebook is used throughout. NH₃ emissions from animal husbandry are assessed using the GAS-EM procedure (Dämmgen et al., 2002a), which reflects the detailed methodology of the future third edition of the

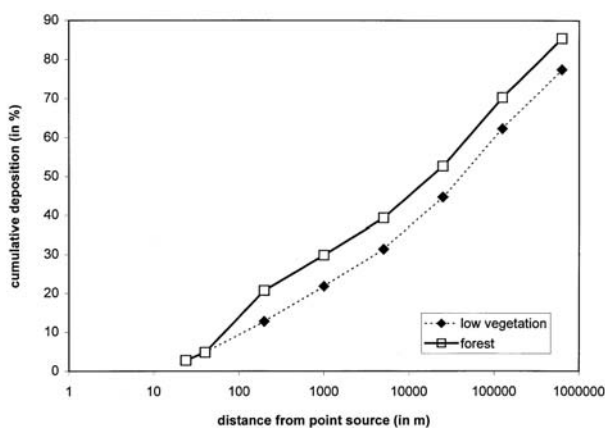


Fig. 4
Cumulative NH₃ deposition as a function of distance from point source (after Erisman, 2000)

EMEP/CORINAIR guidebook. For all other categories including human beings, the simpler methodology of the second edition of the EMEP/CORINAIR guidebook is used. The non-agricultural ammonia emissions calculated for Germany and 1999 add up to 78.4 Gg according to Handley et al. (2001), which would be 12 % of the German national total. Non-agricultural sources to be considered - sorted according to their contribution - are transport, sewage sludge, domestic heating of solid fuels, pets and fertilizer production.

In the detailed approach for animal husbandry, the fate of nitrogen is depicted from the animals' mouth to the soil, considering feeding, animal performance, housing and grazing, storage and application. Each step for each animal category is characterized by a variable partial emission factor, which is deduced from frequency distributions of the various management types. These have to be assessed by questioning experts in each district (Landkreis). The results obtained from respective districts will be used to feed the RAUMIS sector model (Döhler et al., 2002), which generates frequency distributions for all relevant activities.

The quantification of biogenic emissions of nitrogen oxides (NO, N₂O) according to the current EMEP/CORINAIR guidebook is inadequate. It has to be replaced by dynamic modelling such as described by Li et al. (2000) and Stange et al. (2000).

2.2 Modelling atmospheric transport and deposition

There is a difference between deposition modelling and long-range transport modelling, i.e. the focus of the two is different: deposition modelling is aimed at estimating the input to ecosystems as accurately as possible; long-range transport modelling, on the other hand, is aimed at determining the distribution of pollutants (concentration and deposition) based on emission estimates and dispersion calculations. In long-range transport modelling, deposition is a loss term in which the accuracy needed is dependent on the modelled scale. When both the pollution distribution and the deposition need to be known as accurately as possible, a compromise is required on the detail: some of the aspects will be modelled in order to take the main processes determining transport and deposition into account. In applying abatement strategies on a national or European scale, it is desirable to know both the deposition and the distribution of pollution as accurately as possible. When emission abatement strategies are based on the critical load concept, the deposition has to be determined on the ecosystem level. Desired emission reductions based on the target levels should be known per country and/or per individual source or activity. Therefore, long-range transport modelling and ecosystem-scale deposition modelling should be linked.

The deposition model used here to determine the deposition in Hesse is the EDACS model (Estimation of Deposition of Acidifying Components in Europe). Wet deposition in these models is estimated using interpolation of wet deposition measurements. Dry deposition is determined using the inferential technique (Hicks et al, 1987; Erisman and Draaijers, 1995). The dry deposition flux is inferred from interpolated concentrations and land-use-specific dry deposition velocities. The resistance framework used in the inferential technique is extensively described in Erisman and Draaijers (1995).

The outline of the EDACS model to estimate local and regional scale deposition fluxes is presented in Fig. 5. The basis for the two estimates is formed by calculations with the EMEP long-range transport model. With this model dry, wet and total deposition is estimated on a 50 x 50 km² grid over Europe using emission maps for SO₂, NO_x and NH₃ (Vestreng and Støren, 2000). The model results are used for estimating country-to-country budgets, as a basis of sulphur and nitrogen protocols, and for assessments. The local-scale approach used by ECN depends strongly on LTRAP model results. Calculated ambient concentrations of the acidifying components (daily averages) and concentrations in precipitation (monthly averages) are used along with a detailed land-use map and meteorological observations to estimate small-scale dry deposition fluxes (Fig. 5). By using calculated concentration maps, the relationship between emissions and deposition is maintained and scenario studies, budget studies and assessments can be carried out on different scales. Wet deposition is added to the dry deposition to estimate total local scale deposition in Europe. Wet deposition can either be obtained directly from the EMEP model, or from measurements made in Europe. The advantage of the EDACS type of modelling is that they are independent of the LTRAP model that is used as input. Either concentration

measurements or an other model can be used. For the application in Hesse the OPS model will be used to generate the concentration fields on a 5 x 5 km² scale and wet deposition maps will be derived from interpolation of measurements. For these calculations emission maps of NO_x, SO₂ and NH₃ on a 5 x 5 km² resolution are necessary. The large-scale background concentrations can be obtained using the EMEP model results or OPS.

2.3 Experimental validation of modelled concentrations and depositions

The spatial and temporal patterns of the above mentioned emissions and the respective meteorological parameters influence the atmospheric concentration fields, the atmospheric chemical reactions and transport phenomena which together with the local surface properties govern the deposition. For ecosystems, the deposition density is the only adequate dose in dose-response relationships (Dämmgen et al., 1997).

The parameters available at air quality monitoring stations located in rural and forested environments (see Fig. 6) are listed in Table 2.

The resolution in time of all measurements except N₂O and methane (CH₄) concentrations is 30 min. N₂O and CH₄ concentrations are determined with gas chromatography every fourth day with a resolution of approx. 30 min.

The official air quality monitoring stations in Hesse are not equipped with samplers or monitors for concentration measurements of NH₃, HNO₃ and the respective aerosols constituents, as automated detection and collection equipment is not available. In principle, the simultaneous determination of the reactive gases and their reaction products is likely to be biased if gases are not stripped from the air stream prior to the collection of the aerosols. The aerosols themselves are thermally unstable; therefore measures have to be taken to collect their dissociation products. Only denuder-filter combinations meet the requirements of comprehensive concentration measurements (cf Dämmgen 2001a). The best monitoring device to be used for the online measurements of gases and aerosol components is the Steamjet Aerosol Collector (Sjac; Slanina et al., 2001). The Sjac uses rotating denuders to strip the gas and steam injection to grow and collect the aerosols. The solutions are pumped to an IC analyser and are analysed for the major inorganic components. In this way hourly data for the gases NH₃, SO₂, HNO₃, HONO and HCl and the aerosol components NH₄⁺, SO₄²⁻, NO₃⁻ and Cl⁻ can be obtained. The equipment is relatively expensive, but yields hourly data with a good accuracy.

Within this programme, mean concentrations of the reactive N species, their concentration partners and products (NH₃, HNO₂, HNO₃, SO₂, HCl; as well as aerosol NH₄⁺, NO₃⁻, SO₄²⁻ and Cl⁻) are determined with KAPS (Kananaskis Air Pollutant Sampler: Peake, 1985; Peake

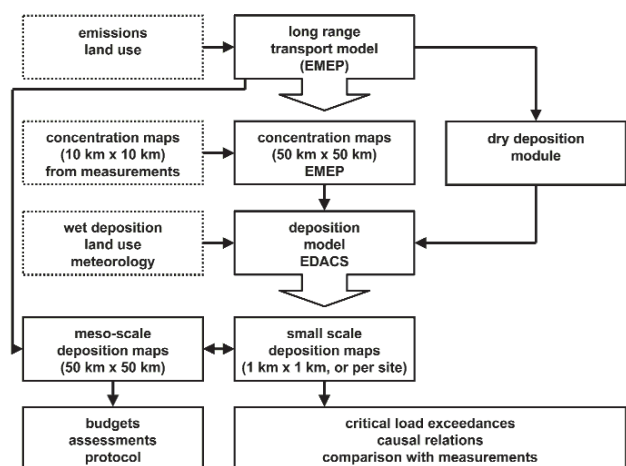


Fig. 5
Outline of the system to model deposition

Table 2
Meteorological parameters and trace gases measured at the standard air quality monitoring stations of Hesse

<i>meteorological parameters</i>	standards applied	Stations
- wind velocity and direction	VDI 3786 sheet 2	all stations except Zierenberg
- air temperature at z = 2 m above ground	VDI 3786 sheet 3	all stations
- soil temperatures, various depths	VDI 3786 sheet 13	Linden
- air humidity at z = 2 m above ground	VDI 3786 sheet 4	all stations
- air pressure at z = 2 m above ground	VDI 3786 sheet 16	Frankenberg, Fürth/Odenw., Königstein, Spessart, Witzenhausen
- global radiation	VDI 3786 sheet 5	all stations except Zierenberg
- precipitation (Hellmann sampler)	VDI 3786 sheet 7	Linden
<i>trace gas concentrations at z = 3.5 m above ground</i>		
- sulphur dioxide (SO ₂)	DIN EN 14212	all stations
- carbon monoxide (CO)	VDI 2455 sheet 2	Linden, Zierenberg
- carbon dioxide (CO ₂)	---	Linden
- nitric oxide (NO)	VDI 2453 sheet 3	all stations
- nitrogen dioxide (NO ₂)	VDI 2453 sheet 3	all stations
- non-methane hydrocarbons (C _n H _m)	VDI 3483 sheet 2	Riedstadt (C _n H _m), Linden (BTX)
- ozone (O ₃)	VDI 2468 sheet 6	all stations
- nitrous oxide (N ₂ O)	---	Linden
- methane (CH ₄)	---	Linden

and Legge, 1987) at the Environmental Monitoring and Climate Impact Research Station Linden (Grünhage et al., 1996) with a resolution in time of one week separately for day and night hours. For details see Zimmerling et al. (2000a).

Spatial and temporal variability of HNO₂ and HNO₃ concentrations can be estimated using a combined photochemistry, transport and deposition model. Such a model is currently not available.

Because of the high spatial variability of NH₃ concentrations a small passive sampling network (Figure 5) using batch type samplers as developed by Ferm (1991) was established to determine mean concentrations (Dämmgen 2001b). The resolution in time is one month.

As mentioned above, NH₄⁺, NO₃⁻, SO₄²⁻ and Cl⁻ in aerosols are determined using KAPS at the Linden site. The cut-off point is approx. 3 µm. With regard to the frequency distributions of particle diameters, this is in practice equal to PM_{2.5} (cf Finlayson-Pitts and Pitts, 1986). The spatial variability of the concentrations of aerosol constituents in regions not influenced by strong point sources is comparatively small (Zimmerling et al., 2000b). Thus, from the measurements at Linden station we will estimate concentrations fields using additional information provided by concentration measurements of total suspended matter and PM₁₀ (DIN EN 12341), respectively, at the sites given in Fig. 6.

Deposition of sedimenting particles is measured by use of surrogate surfaces. The existing network serving the needs of forest ecosystem research and operated by the Hessische Landesanstalt für Forsteinrichtung, Waldforschung und Waldökologie using Münden samplers (HLfU, 1986) is supplemented by a new dense network



Fig. 6
Locations of ambient air monitoring stations for trace gases and meteorological parameters (circles), of aerosol monitoring stations (triangles) and of the passive sampling network for ammonia (squares) located in rural and forested environments



Fig. 7
Locations of bulk deposition sampling stations (HLUG monitoring network - Rotenkamp samplers, circles; Hessian forest monitoring network - Münden samplers, squares)

using Rotenkamp samplers (Dämmgen et al, 1994b; Dämmgen et al., 2000). For locations see Fig. 7.

2.4 Time frame

The project described commenced in 2000. First measurements in addition to the official Hessian air quality and forest monitoring networks began in 2001 (passive sampling of NH_3 , HLUG bulk deposition, denuder-filter measurements). Detailed emission inventories are being prepared 2002/2003.

Because emission inventories depict typical emissions rather than actual emissions i.e. they do not reflect actual climate and weather conditions and farm management according to actual weather conditions, mean concentration and mean deposition fields have to be obtained from measurement over several years, the representativity of which has to be proved by adequate time series of meteorological, concentration and deposition data sets.

Atmospheric transport and chemistry as well as deposition modelling which has to rely on emission inventories

and has to be validated by solid experimental data, is projected for 2004.

Additional effort will be necessary to establish knowledge about typical gradients above the major types of forests. Such “tower” measurements have to be combined with measurements describing the physiological activity of the ecosystem (e.g. vertical CO_2 and H_2O fluxes). Because of the importance of aerosol deposition to forests (Table 1) further attention has to be paid to experimental verification of the model assumptions for aerosol deposition with regard to N species. At present, both gradient and relaxed-eddy accumulation techniques are being developed for remote operation. The time-averaged gradient technique using sampling techniques seems to be adequate.

Modelling HNO_3 concentration fields relies on sufficient knowledge of the HNO_3 formation processes which are closely related to O_3 dynamics. Simultaneous monitoring of HNO_3 and O_3 concentrations and vertical fluxes in comparatively remote areas and such areas which are at present at risk due to increased deposition and atmospheric chemistry is therefore desirable. The Linden station can be thought to be representative of large parts of central and northern Hesse. The different physical and pollution climates of southern Hesse require a further extension or completion of an air monitoring station.

Combined transmission and regional deposition models with a high resolution in space and time are presently developed within the BIATEX scientific community (Erisman et al., 2002).

The present state of the project is documented under <http://www.uni-giessen.de/~gf1034/DEUTSCH/-DUAH/-DUAH.htm> which will be updated regularly. It also describes the materials and methods used in detail.

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