

# Monitoring and modelling of biosphere/atmosphere exchange of gases and aerosols in Europe

Jan Willem Erisman<sup>a,\*</sup>, Alex Vermeulen<sup>a</sup>, Arjan Hensen<sup>a</sup>, Chris Flechard<sup>a</sup>,  
Ulrich Dämmgen<sup>b</sup>, David Fowler<sup>c</sup>, Mark Sutton<sup>c</sup>, Ludger Grünhage<sup>d</sup>,  
Juha-Pekka Tuovinen<sup>e</sup>

<sup>a</sup>Energy Research Centre of the Netherlands (ECN), P.O. Box 1, 1755 ZG Petten, The Netherlands

<sup>b</sup>Federal Agricultural Research Centre, Institute of Agroecology, D-38116 Braunschweig, Germany

<sup>c</sup>CEH, Bush Estate, Penicuik, Midlothian EH26 0QB, UK

<sup>d</sup>Institute for Plant Ecology, Justus-Liebig-University, D-35392 Giessen, Germany

<sup>e</sup>Finnish Meteorological Institute, FIN-00810 Helsinki, Finland

Received 8 January 2004; accepted 2 July 2004

*Monitoring and modelling of the deposition of sulphur and nitrogen components and the exposure of ozone has gained much progress through the research within BIATEX.*

## Abstract

Monitoring and modelling of deposition of air pollutants is essential to develop and evaluate policies to abate the effects related to air pollution and to determine the losses of pollutants from the atmosphere. Techniques for monitoring wet deposition fluxes are widely applied. A recent intercomparison experiment, however, showed that the uncertainty in wet deposition is relatively high, up to 40%, apart from the fact that most samplers are biased because of a dry deposition contribution. Wet deposition amounts to about 80% of the total deposition in Europe with a range of 10–90% and uncertainty should therefore be decreased. During recent years the monitoring of dry deposition has become possible. Three sites have been operational for 5 years. The data are useful for model development, but also for model evaluation and monitoring of progress in policy. Data show a decline in SO<sub>2</sub> dry deposition, whereas nitrogen deposition remained constant. Furthermore, surface affinities for pollutants changed leading to changes in deposition. Deposition models have been further developed and tested with dry deposition measurements and total deposition measurements on forests as derived from throughfall data. The comparison is reasonable given the measurement uncertainties. Progress in ozone surface exchange modelling and monitoring shows that stomatal uptake can be quantified with reasonable accuracy, but external surface uptake yields highest uncertainty.

© 2004 Elsevier Ltd. All rights reserved.

*Keywords:* Atmosphere-biosphere exchange; Gases; Aerosols; Model; Measurements; Wet deposition; Dry deposition

## 1. Introduction

Deposition of air pollutants is an important loss process for pollutants from the atmosphere and can

cause severe damage to ecosystems. The loss from and the emission to the atmosphere are also important for modelling of long-range transport of secondary formed particulate matter. Furthermore, fluxes of ozone are relevant for determining critical thresholds for effects. In order to develop effective emission reduction options and in order to monitor the progress in terms of reduced deposition levels, a combination of modelling and

\* Corresponding author. Tel.: +31 224 564155; fax: +31 224 563488.

E-mail address: [erisman@ecn.nl](mailto:erisman@ecn.nl) (J.W. Erisman).

measurement activities is needed (e.g. Erisman and Draaijers, 1995). The measurements serve as independent checks on the modelled concentration and deposition fields and they serve as understanding of surface exchange processes (e.g. Erisman et al., 2001). Models serve to keep the link with emissions and to assess effective abatement strategies (e.g. Tarrason and Schaug, 2000). Furthermore, there is a need for tools to upscale results to sites where no measurements are available (e.g. Erisman et al., 1998a). Deposition can occur as wet, dry and cloud/fog deposition. Wet deposition in Europe is routinely monitored in existing networks, e.g. ECE-EMEP (Schaug et al., 1993); this is not the case for dry and cloud/fog deposition of gases and particulate matter, which is much more difficult to measure. On average in Europe, dry deposition accounts for about 20% of the total deposition with a range of 10–90% (Erisman et al., 1998a). Dry deposition of gases and aerosols is site specific, determined by factors such as roughness of the surface, surface wetness, climate and environmental factors. Total (throughfall) deposition in forests is routinely measured at more than 400 intensive monitoring sites of the EU/ICP Forest monitoring programme (De Vries et al., 2001). Apart from these monitoring programmes, several EU projects and EUROTRAC activities have contributed to the needs of monitoring and modelling of surface exchange of trace gases and particles. In this paper a (limited) overview of the monitoring and modelling results and its evaluation are given with the aim of identification the current monitoring and modelling needs.

## 2. Wet and bulk deposition of sedimenting particles

### 2.1. Measuring wet and bulk deposition

Wet and bulk deposition is routinely monitored at about 90 EMEP sites distributed over Europe and in several national monitoring networks [e.g. Germany (Brandenburg: Dämmgen and Zimmerling, 2002; Hesse: Grünhage et al., 2002), UK (Smith et al., 2000), The Netherlands (Erisman and Draaijers, 1995)]. Van Leeuwen et al. (1996) compiled a wet deposition map of Europe based on these measurements for the year 1995. Since then this exercise has not been repeated.

At Schagerbrug, a grassland site close to the Dutch shore, a comparison experiment was conducted of bulk and wet-only samplers used in different national and EU monitoring networks (Erisman et al., 2003). More than 90% of the samplers used are bulk samplers collecting wet deposition and dry deposition of sedimenting particles on the funnels. These results therefore exceed the wet deposition flux by 5–40%, depending on location, climate, sampler construction and chemical component. The results show that for bulk precipitation

fluxes the required accuracy of 10% or less is realised for only 5–40% of the samplers, depending on chemical component. Bulk precipitation fluxes of  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{H}^+$  and Kjeldahl-N generally could be determined with a larger accuracy than bulk precipitation fluxes of  $\text{SO}_4^{2-}$ ,  $\text{Na}^+$ ,  $\text{Cl}^-$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ , Alkalinity and  $\text{H}^+$ . Bulk precipitation fluxes of  $\text{K}^+$  generally had the lowest accuracy. These results show that wet deposition, the dominant input to ecosystems, is not determined accurately enough for trends. An extensive discussion of the factors influencing wet and bulk deposition of sedimenting particles is given in Dämmgen et al. (2004a). The most important factors determining the uncertainty are: (i) the influence of dry deposition of gases and aerosols on the funnels of the samplers; (ii) construction and placement of the samplers; and (iii) sample handling and analysis. Apart from the dry deposition to the funnels, Zimmermann has identified all the factors described in Dämmgen et al. in the first paper on requirements of wet deposition sampling in 1825 (Zimmermann, 1825)!

### 2.2. Modelling wet deposition

Modelling of wet deposition is done for years in a very simple way by using scavenging coefficients together with precipitation amounts (e.g. Tarrason and Schaug, 2000). This approach suffices for annual average wet deposition inputs over large areas or if the annual average loss of particles and gasses by wet deposition needs to be known. Models can estimate the annual average wet deposition with an accuracy of about 25% (EMEP/CCC, 1996) for sulphur, oxidised nitrogen, base cations reduced nitrogen. For reduced nitrogen the contribution of local sources to the wet deposition can be large in areas with high ammonia emission intensities, resulting in large spatial variation in fluxes (e.g. Erisman and Draaijers, 1995). Recently there is a development to more process-based models to describe the formation of clouds and the precipitation processes (e.g. Jung et al., 2003; Loosmore and Cederwall, 2004). These models can describe event based wet deposition fluxes but are still inadequate to describe annual average fluxes over Europe.

## 3. Dry deposition of gases and aerosols

### 3.1. Monitoring dry deposition

Several long-term flux monitoring stations for  $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{O}_3$  and  $\text{NH}_3$  have been established in Europe (Erisman et al., 2001). Measurements were performed at three core stations: Auchencorth (Scotland), peat bog vegetation 10 cm high in semi-background conditions; Speuld (The Netherlands), a forest site with pine trees of

about 20 m height in a polluted area; and *Melpitz* (Germany), a grassland site with vegetation height of about 15 cm, also in a source region. On all locations measurements are performed in towers. Vertical gradients of the concentrations of  $\text{SO}_2$ ,  $\text{NO}_x$  and  $\text{NH}_3$  are continuously measured, together with meteorological data by a sonic anemometer. The change of the concentration with height is used to calculate the deposition flux. Such measurements have provided the major progress in understanding the processes, which regulate long-term surface-atmosphere fluxes. The three sites (Speulder forest in The Netherlands, Auchencorth in Scotland and Melpitz in Germany) represent different regions in Europe to estimate local inputs and to validate deposition models, which are currently used or developed to estimate ecosystem specific deposition in Europe. Fluxes at Auchencorth Moss are lowest for all components, except for those components much influenced by the sea as a source. As Melpitz is located far away from seas, these components are lowest at this site. Wet deposition is the dominant source of input at Auchencorth, whereas at Speulder forest, through its roughness and pollution climate, dry deposition is dominant. At this site dry deposition velocities are highest. Melpitz is a polluted site. Particularly sulphur deposition is high. Four years of continuous measurements are now available, and a first trend analysis is now possible; over 4 years this shows for example the decrease in the  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  levels at all sites. For  $\text{NH}_3$  and  $\text{NO}_x$  no significant trend was detected. The data is also used to develop dry deposition parameterisations. The continuous flux measurements in the moorland ecosystem (Auchencorth Moss) in Scotland were used to explore the processes governing external plant deposition (Fowler et al., 2001). By comparing the data from the three sites it was shown that the dry deposition of  $\text{SO}_2$  is influenced by  $\text{NH}_3$ , because the surface resistance ( $R_s$ ) shows a dependence on the molar ratio of  $\text{NH}_4$  over  $\text{SO}_4 + \text{NO}_3$  in precipitation. Furthermore, long-term data at Speuld show that deposition monitoring is necessary because surface affinities can change, e.g. due to meteorological conditions. It is therefore necessary to monitor the effect of emission reductions on a number of stations in Europe.

Any vertical flux density determined in the atmospheric surface layer which is to reflect the spatially representative exchange at the surface of the system considered requires this layer to be stationary and horizontally homogeneous. Suitable tools to estimate the influence of a limited horizontal extension of the system under investigation are the two-dimensional source-area approach (cf. Schmid, 1994) or the one-dimensional footprint approach (cf. Horst and Weil, 1992; Horst, 1997, 1999; Haenel and Grünhage, 1999, 2001; Kormann and Meixner, 2001). In addition, they have to represent in space the typical atmospheric

conditions governing the fluxes. Due to financial restrictions, these measurements are normally point measurements. Therefore, the spatial variability and the typical deviations have to be characterized. Within BIATEX-2, several studies have been performed to quantify deviations of meteorological parameters (global radiation, air temperature, horizontal wind velocity), driving forces (vertical gradients of air temperature and horizontal wind velocity) and vertical fluxes of sensible and latent heats, momentum and carbon dioxide (e.g. Dämmgen et al., 2001, 2002, 2004b; Grünhage and Haenel, 2001).

#### 4. Modelling dry deposition

Modelling dry deposition is dependent on the spatial resolution needed. Whereas local scale Soil-Vegetation-Atmosphere-Transfer (SVAT) models rely on the detailed description of the energy balance of the systems considered, regional or national scale models make use of simplifying or integrating assumptions and use typical deposition velocities rather than site-specific driving forces, such as compensation points or actual stomatal resistances. SVAT models serve two purposes: in agricultural and forest meteorology they are a tool to calculate water dynamics, e.g. to predict irrigation or other management measures. In forests, prediction of ground water formation or of run-off may be essential. In the context of the ecotoxicology of air pollutants they are needed to describe flux effect relationships. In ecosystems, the flux relevant for effects of acidification is the overall input of acidifying species into the system as a whole. For ozone, the toxicologically relevant flux is the partial flux through the stomata—the flux to external surfaces is toxicologically almost irrelevant under ambient conditions in Europe. Therefore, the modelling of stomatal behaviour is crucial for the establishment of dose–response relationships (Dämmgen et al., 1997; Grünhage et al., 2004; Tuovinen et al., 2004). SVAT models for this purpose have to be validated independently of the respective air pollutant using water vapour or carbon dioxide exchange measurements.

##### 4.1. Nitrogen and sulphur compounds

Within BIATEX-2, SVAT models have been developed in particular for the description of the soil/vegetation-atmosphere exchange of ammonia (Sutton et al., 2001). Erisman et al. (1997) developed a SVAT model for aerosol deposition. These models have been validated independently, e.g. for PLATIN within the VERTIKO experiments for various vegetation types (arable land, forest; Schaaf et al., unpublished results). The big-leaf model PLATIN has been used to estimate

fluxes of  $\text{NH}_3$ ,  $\text{HNO}_2$ ,  $\text{HNO}_3$  and  $\text{SO}_2$  between the atmosphere and forest and cropped ecosystems (Grünhage et al., 1998; Dämmgen and Zimmerling, 2002).

The development of models used to calculate regional and site-specific inputs of sulphur and nitrogen have relied heavily on the long-term measurements at Melpitz, Speulder Forest and Auchencorth Moss. The measurements show that  $\text{SO}_2$  fluxes over most of Europe are regulated by chemical processes within surface films of water, which in turn is strongly influenced by the presence of  $\text{NH}_3$ . Dynamic models, which simulate the process, have been developed and where sufficient chemical and micrometeorological data are available, these models quantify the net exchange of  $\text{SO}_2$  and  $\text{NH}_3$  (Flechard et al., 2000). Fig. 1 shows a comparison of different surface exchange parameterisations with measured fluxes (Auchencorth, Scotland) by Flechard et al. (2000). The dynamic model provides somewhat better results than the two different static resistance models, which are empirical models derived from the (limited) set of observations (e.g. Erisman and Draaijers, 1995). The major improvement is expected in periods where there are big changes in the concentration ratios of the different components. In those cases the ‘chemical memory effect’ of the surface that to a large extent determines the flux is taken into account.

For  $\text{NH}_3$  parameterisations of the bi-directional plant-atmosphere exchange and the influence of management options have been included in the models (Sutton et al., 1998, 2001). These past years several monitoring studies and joint field experiments have been conducted to improve our understanding of the atmosphere–biosphere exchange of ammonia (see Sutton et al., 1996, 1998, 2001 for overviews). The main emphasis has been on the description of the plant–atmosphere interactions and the stomatal flux of ammonia, which can now be described quite reasonable (Nemitz et al., 2001; Sutton et al., 2001). The external surface parameterisations can potentially be described by the dynamic models, but these are not ready to be

used in models used for generalisation. The reason for this is that these long-range transport models the emissions are taken as inputs and are therefore modelled separately from the deposition. A bi-directional description of the surface exchange process needs large modifications of the long-range transport models. The dynamic models can be used in inferential models without much modification.

Within BIATEX-2 models to describe site-specific deposition and the fluxes within Europe on a local/regional scale have been updated. The parameterisation of the dry deposition velocity for gases and particles was based on Erisman and Draaijers (1995) and was further developed using recent literature (e.g. Brook et al., 1999; Wesely and Hicks, 2000) and results from BIATEX-2 (Sutton et al., 2001; Erisman et al., 2001; Fowler and Erisman, 2003). The models have been applied over European scales for annual deposition estimates of sulphur and nitrogen inputs (see below).

Long-term measurements are also valuable for evaluation of policies. A clear example of the need for monitoring is obtained from the long-term flux measurements at Speulder forest. Fig. 2 shows the change in deposition parameters between 1993 and 1998. It shows that until 1997 the flux followed the decrease in concentration. At the same time the  $R_c$  values (expressed as the years median value) increased, probably because the flux of  $\text{NH}_3$  (also displayed in Fig. 1) decreased. The flux of  $\text{NH}_3$  is important because a higher flux stimulates the uptake of  $\text{SO}_2$  at the surface due to the co-deposition effect (Adema et al., 1986; Erisman and Wyers, 1993). After 1997 the  $\text{NH}_3$  flux increased, leading to a decrease in the  $R_c$  of  $\text{SO}_2$ . This is clearly a support for dynamic modelling.

#### 4.2. Ozone

For  $\text{O}_3$  the models provide the potential to be used for flux-based approach for application of a critical loads methodology, to replace the AOT40 concept

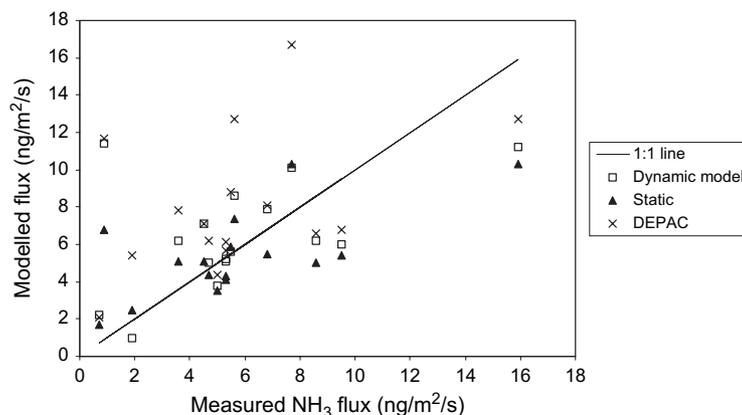


Fig. 1. Comparison of different surface exchange parameterisations (based on data from Flechard et al., 2000).

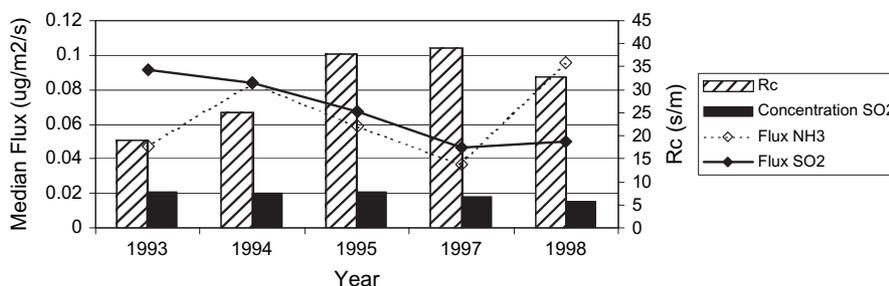


Fig. 2. Change in annual concentration, flux and surface resistance of SO<sub>2</sub> measured at Speulder forest.

(Grünhage and Haenel, 1997, 2000; Tuovinen et al., 2004). Up to now ozone fluxes were assumed to be determined largely by stomatal uptake. As stomatal control is generally described in relation with water vapour or photosynthesis, parameterisations of stomatal resistance in models can be validated via water vapour (e.g. Grünhage and Haenel, 1997) or CO<sub>2</sub> flux measurements. There are much more uncertainties with respect to the parameterisation of O<sub>3</sub> deposition on external plant surfaces. Many studies illustrate that non-stomatal O<sub>3</sub> deposition can be an important part of total deposition (e.g. Fowler et al., 2001; Simpson et al., 2001). Fowler et al. (2001) found at Auchencorth Moss that the inferred canopy resistance due to non-stomatal O<sub>3</sub> deposition decreases exponentially with increasing solar radiation (Fig. 3). The interpretation is that the non-stomatal flux represents thermal decomposition of O<sub>3</sub> at the surface.

Generally, vertical flux densities of O<sub>3</sub> are influenced by (photo)chemical reactions in the surface layer, e.g. by reactions with NO emitted from the soil and in forest ecosystems by reactions with volatile organic compounds emitted from the trees. Especially in coniferous forests, O<sub>3</sub> deposition velocity during night-time hours is approximately half of that during the days (cf. Pilegaard, 2001). Recently, Ludwig et al. (2001) published a parameterisation of biogenic NO emissions

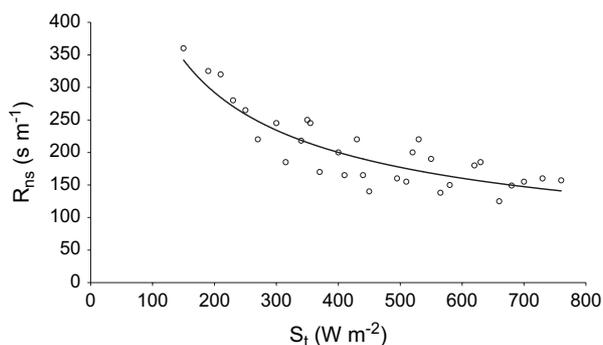


Fig. 3. The relationship between canopy resistance due to non-stomatal O<sub>3</sub> deposition ( $R_{ns}$ ) and solar radiation ( $S_t$ ) for Auchencorth Moss daytime data, dry surfaces (source: Fowler et al., 2001, Fig. 5).

for different land-use classes and mean climatological mid-European conditions. This approach might increase the accuracy of NO flux estimates and their influence on O<sub>3</sub> deposition in regional-scale modelling.

## 5. Total deposition

### 5.1. Monitoring

Currently, throughfall measurements are made at about 400 Intensive Monitoring plots to estimate the site-specific inputs of nutrients in European forests (De Vries et al., 2001). Throughfall data are a measure for the soil load of pollutants. Because of canopy exchange processes and transport in the plant phloem these data cannot directly be used for atmospheric deposition (Erisman and Draaijers, 1995; Dämmgen et al., 1997). The measurements are, however, relatively cheap and easy to perform, and are therefore suitable to obtain spatial variation in deposition.

### 5.2. Modelling

Canopy exchange models have been developed to overcome this problem (Ulrich, 1983; Draaijers and Erisman, 1995; Draaijers et al., 1996). By applying such models an estimate of total deposition can be obtained, but the uncertainty is still relatively high. Fig. 4 shows the deposition of nitrogen to forests in Europe where throughfall is measured within the Pan European monitoring programme.

Throughfall measurements can be used for detection of trends in deposition. Fig. 5 shows the results of long-term measurements at Speulder forest in the centre of the Netherlands. It includes both the deposition derived from throughfall measurements and from micro meteorological measurements. The emissions are for the province in which the Speulder forest is situated. Despite the large scatter between the years, mainly determined by meteorological variation, the deposition estimates represent the trend in emissions for the area.

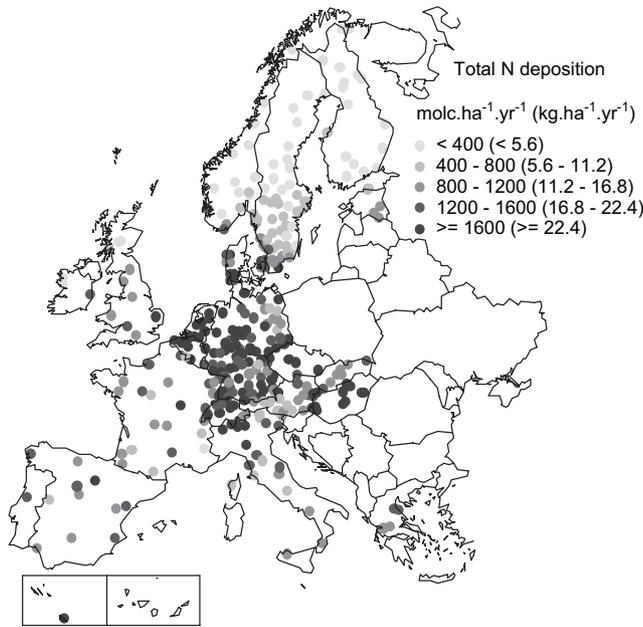


Fig. 4. Nitrogen deposition to forests in Europe ( $\text{mol ha}^{-1} \text{y}^{-1}$ ).

**6. Modelling of deposition of sulphur and nitrogen compounds**

There are two approaches to model ecosystem inputs: (i) long-range transport modelling based on emission estimates and (ii) using measurements of concentrations and meteorological data to estimate deposition, or a combination of the two. Within BIATEX-2 the

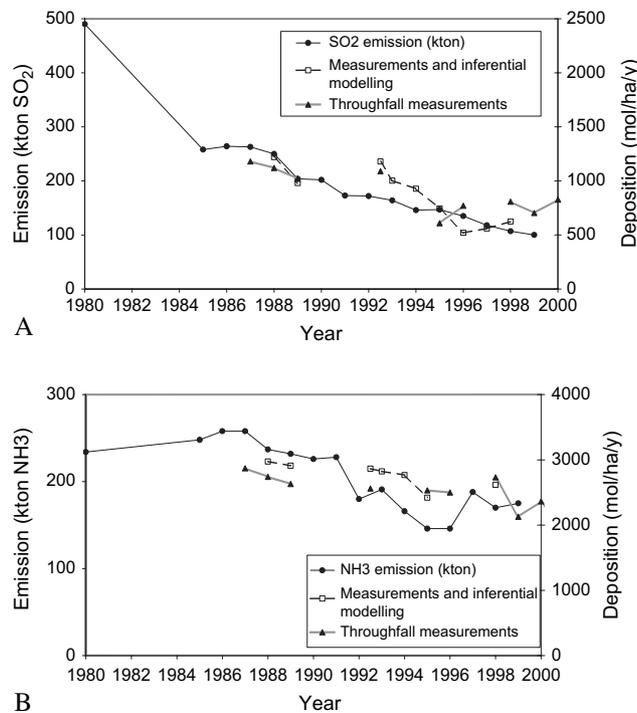


Fig. 5. Long-term deposition measurements of  $\text{SO}_4$  (A) and  $\text{NH}_4$  (B) at Spouder forest in the Netherlands. ( $\text{mol ha}^{-1} \text{y}^{-1}$ ).

combination of the two has been chosen to maintain the link with emissions and at the same time obtain ecosystem-specific deposition estimates. A tool to calculate site-specific deposition fluxes, EDACS (European Deposition of Acidifying Components on a small Scale) was developed at RIVM (Erisman and Draaijers, 1995). This model is now reprogrammed and improved by ECN and meteorological input is updated, based on data from the European Centre for Meteorology and Weather Forecast (ECMWF). Concentrations of gases and aerosols are derived from the EMEP model or from the DEHM model (Brandt et al., 2001). Wet deposition is derived from measurements or from the EMEP/DEHM model and dry deposition is estimated using modelled concentrations with the EMEP model and land use specific dry deposition velocities calculated with ECMWF meteorology and a dry deposition module (DEPAC) (Fig. 6). Every 6 h the ECMWF meteorological data is used to calculate a deposition velocity for each grid or each plot. The deposition velocity is combined with a concentration to estimate the flux. An annual flux is the summation of all 6-h values. The model input and output is flexible and depends on the land use information that is used. Currently land use maps for Europe ( $1/6^\circ \times 1/6^\circ$ ) and for different countries ( $1 \times 1 \text{ km}^2$ ) are available. 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. An example of EDACS outcome is given in Fig. 7, where total deposition of  $\text{SO}_x$  in Germany is plotted for the years 1993 and 1999.

In this approach we made a site-specific calculation using site-specific information available for the Intensive Monitoring plots. The EMEP model results are only available until 1996. Since then, a new model was developed and is now made operational. The comparison

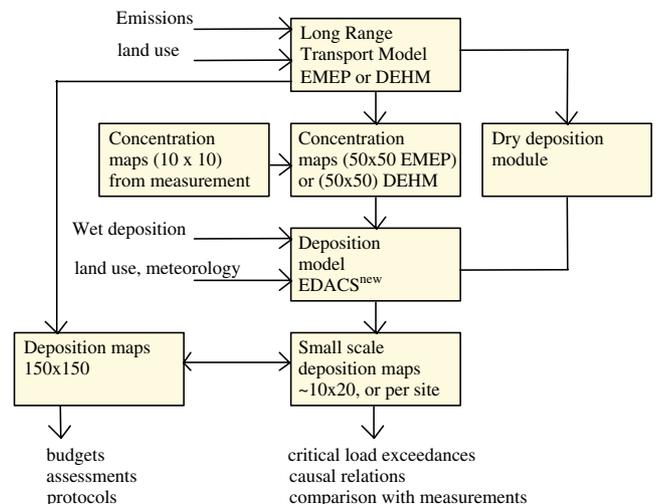


Fig. 6. Outline of method to estimate local scale deposition fluxes.

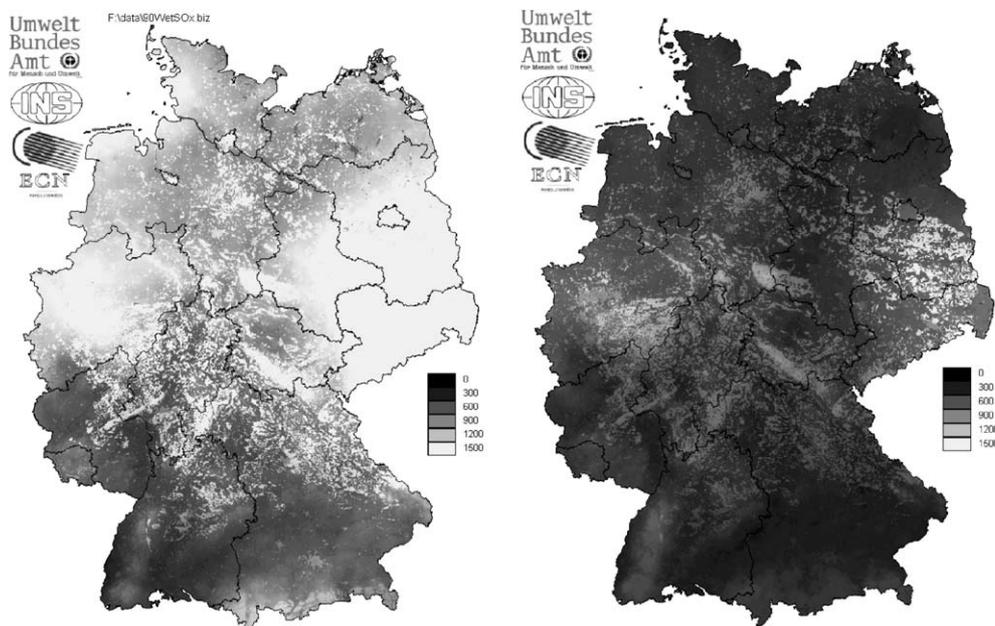


Fig. 7. Dry deposition of  $SO_x$  in Germany ( $1 \times 1 \text{ km}^2$ ) as calculated with EDACS for 1990 (A) and 1999 (B) ( $\text{mol ha}^{-1} \text{y}^{-1}$ ).

of model results with throughfall data has therefore been made for 1996 (223 plots). Fig. 8 shows the model comparison with throughfall measurements at the plots. The correlation for sulphur is small (Fig. 8A). This is especially due to an overestimation of sulphur deposition at a few sites. These sites are all located close to or in the black triangle. It appears that the EMEP model overestimates  $SO_2$  concentrations in these areas. On average the modelled S deposition and measured throughfall are comparable, with a bias at high throughfall and/or high model estimates. This might be due to local influences, uncertainty in stand characteristics for modelling and/or uncertainty in modelled values. A best regression estimate was

$$SO_{4,\text{model}} = 530 + 0.63 \cdot SO_{4,\text{throughfall}} \quad R_{\text{adj}}^2 = 0.32 \quad (1)$$

The N deposition, both of  $NO_3$  and  $NH_4$  are considerably larger than the measured throughfall, although the correlation is better than for  $SO_4$  (Fig. 8B–D). Specifically the reduced N deposition is higher up to a factor of two, despite the high correlation. Best regression estimate were:

$$NO_{3,\text{model}} = 540 + 0.75 \cdot NO_{3,\text{throughfall}} \quad R_{\text{adj}}^2 = 0.37 \quad (2)$$

$$NH_{4,\text{model}} = 610 + 1.54 \cdot NH_{4,\text{throughfall}} \quad R_{\text{adj}}^2 = 0.64 \quad (3)$$

The overestimation of the model might be the result of the uncertainty in the correction for canopy uptake to calculate the deposition from throughfall measurements.

### 6.1. Modelling of ozone fluxes

A lot of progress in  $O_3$  deposition modelling has been made based on long-term atmosphere-biosphere exchange measurements (see Tuovinen, 2000; Tuovinen et al., 2001 for an overview). This is especially relevant for the flux-based approaches which have been identified as an option for the more advanced ('level II') critical levels, to replace (Emberson et al., 2000) or modify (Grünhage et al., 1999; Tuovinen, 2000) the concentration-based approach employing the AOT40 index. To meet the need to estimate the regional distribution of fluxes to different ecosystems, the photochemical model of EMEP has been improved by introducing a new dry deposition module (Emberson et al., 2001; Simpson et al., 2001; Hole et al., 2004). This module includes a partitioning of the total deposition flux between the stomatal and non-stomatal pathways, the former being the component most closely related to biological effects. The stomatal flux is modelled by using a detailed uptake model that describes the stomatal conductance as a function of phenology and environmental variables for a large number of European plant species. The module can also be used in stand-alone risk assessment applications.

The EMEP ozone deposition module has been tested by comparing predicted deposition velocities and stomatal conductances with observations at both leaf and canopy scale (Emberson et al., 2000, 2001; Tuovinen et al., 2001, 2004; Hole et al., 2004). An example is shown in Fig. 9. The wide range of surface types and climatic regions makes it difficult to model ozone deposition across the whole EMEP modelling

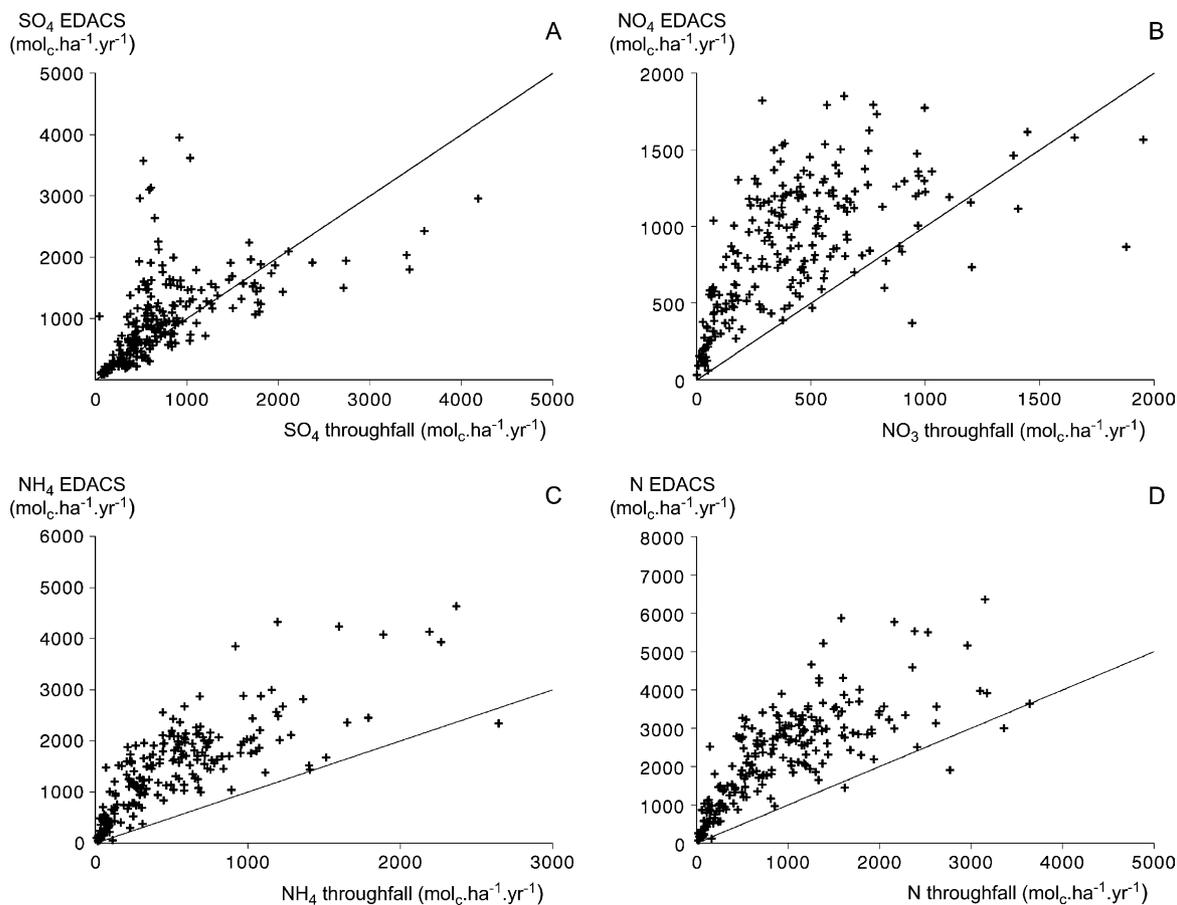


Fig. 8. Comparison of total deposition calculated with EDACS with throughfall measurements of  $\text{SO}_4$  (A),  $\text{NO}_3$  (B),  $\text{NH}_4$  (C) and total N (D) in 1997 and 1998.

domain. However, the comparisons show that the module is capable of producing reasonable results in a variety of conditions. In stand-alone applications local data can be used, which reduces the effect of the variability within the aggregated land-cover classes and uncertainties in regional-scale meteorological data.

## 7. Conclusions

Most of our present knowledge on atmosphere–biosphere exchange stems from the 1980s and early 1990s, when acidification research was intensively conducted. Recently, most progress has been made on  $\text{O}_3$  and  $\text{NH}_3$  exchange and on monitoring of dry deposition fluxes. Examples of the value of deposition monitoring are the non-linearity issue in sulphur emission and deposition and the change in surface affinities and the discovery of the ammonia hole in the Netherlands (Erisman et al., 1998b). Monitoring of fluxes has become possible, but the uncertainty is still very high, especially for wet deposition (40% in the case no standard wet-only samplers are used) and the deposition of reactive gases and particles (>40%).

Wet deposition can be measured with reasonable accuracy (15–20%) if wet-only gauges are used, which are placed within the open field fulfilling the requirements for open field measurements (e.g. Draaijers and Bleeker, 2001). For improvement of dry deposition monitoring there is a need for more robust, sensitive and fast concentration sensors. The gradient methods have been shown to be feasible; however, eddy correlation would provide more robust results and less rejection of data not fulfilling criteria for deposition measurements. New concentration sensors should therefore be developed. Fluxes of ozone and inert gases, such as  $\text{CO}_2$ , can easily be monitored. Progress has been made in the recognition and understanding of  $\text{O}_3$  uptake at surfaces.

For modelling of the surface exchange of gases, there is a lack of parameterisation of dynamic processes, occurring for example with surface wetness. Furthermore, the modelling of the bi-directional nature of gas exchange (nitrogen compounds) and the external surface uptake of  $\text{O}_3$  should be improved. Dynamic models have become available describing the surface wetness chemistry and the bi-directional exchange of nitrogen compounds. These models show better results than the widely used inferential models when compared to

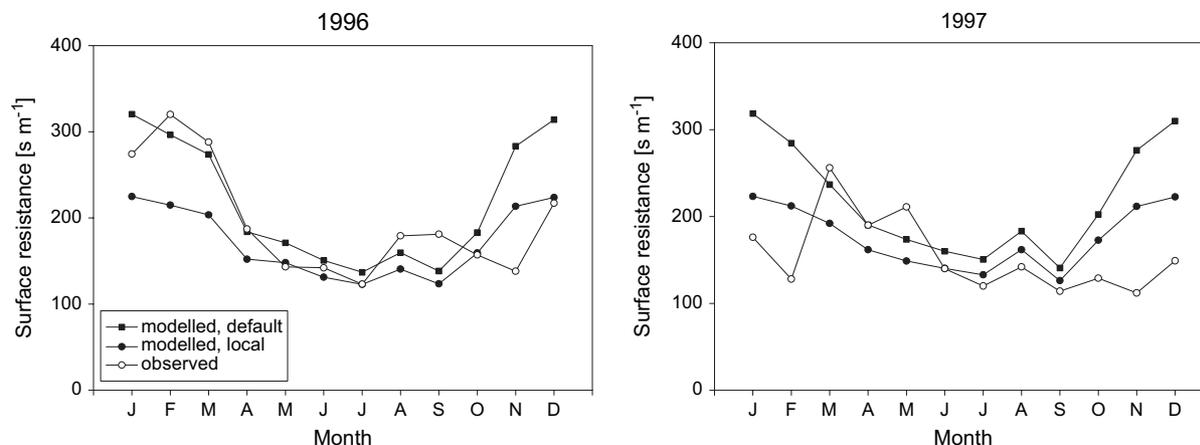


Fig. 9. Measured and modelled total surface resistances during the daytime as monthly medians for a Norway spruce forest at Ulborg, Denmark. The EMEP model calculations have been carried out using both the assumed leaf area index and tree height (default) and the measured values (local). (From Tuovinen et al., submitted)

measurements. However, these models cannot yet be used in long-range transport models because the detailed data needed in the dynamic models is not available, and the emissions are now used as inputs and independently handled from the deposition in the long-range transport models. Therefore, there is a need to improve the long-range transport models first, before the new deposition models can be incorporated. The accuracy of the results obtained greatly depends, furthermore, on the errors in the input parameters as well as on the model itself. Small-scale spatial variability and the accuracy of input parameters has to be reflected by the iterations within the model (error propagation).

## References

- Adema, E.H., Heeres, P. and Hulskotte, J. (1986). On the dry deposition of  $\text{NH}_3$ ,  $\text{SO}_2$  and  $\text{NO}_2$  on wet surfaces in a small scale wind tunnel. Proceedings of the Seventh World Clean Air Congress, Sydney, Australia, pp.1-8.
- Brandt, J., Christensen, J.H., Frohn, L.M., Palmgren, F., Berkowicz, R., Zlatev, Z., 2001. Operational air pollution forecasts from European to local scale. *Atmospheric Environment* 35S, 91–98.
- Brook, J.R., Zhang, L., Di-Giovanni, F., Padro, J., 1999. Description and evaluation of a model of deposition velocities for routine estimates of air pollutant dry deposition over North America. Part I: model development. *Atmospheric Environment* 33, 5037–5051.
- Dämmgen, U., Zimmerling, R., 2002. Vertical fluxes of air-borne acidifying and eutrofying species in the Schorfheide Nature Reserve in Brandenburg, Germany. *Journal of Applied Botany* 76, 190–202.
- Dämmgen, U., Grünhage, L., Jäger, H.-J., 1997. Description, assessment and meaning of vertical fluxes of matter within ecotopes: a systematic consideration. *Environmental Pollution* 96, 249–260.
- Dämmgen, U., Frühauf, C., Haenel, H.-D., 2001. Small scale spatial variability of air temperature, temperature gradients and heat and momentum fluxes. In: Midgley, P.M., Reuther, M., Williams, M. (Eds.), Proceedings from the EUROTRAC-2 Symposium, 2000. Springer, Berlin, CD-ROM.
- Dämmgen, U., Schaaf, S., Frühauf, C., 2002. Accuracy and spatial representativity of heat flux measurements. In: Midgley, P.M., Reuther, M. (Eds.), Proceedings from the EUROTRAC-2 Symposium, 2002. Margraf, Weikersheim.
- Dämmgen, U., Erisman, J.W., Cape, J.N., Grünhage, L., Fowler, D., 2004a. Assessment of atmosphere/biosphere fluxes of sedimenting particles, in preparation.
- Dämmgen, U., Grünhage, L., Schaaf, S., 2004b. The accuracy and spatial variability of parameters needed to determine vertical fluxes of air constituents. *Journal of Applied Botany*, in preparation.
- De Vries, W., Reinds, G.J., Van der Salm, C., Draaijers, G.P.J., Bleeker, A., Erisman, J.W., Auée, J., Gundersen, P., Kristensen, H.L., Van Dobben, H., De Zwart, D., Derome, J., Voogd, J.C.H., Vel, E.M., 2001. Intensive Monitoring of Forest Ecosystems in Europe. Technical Report, 2001a. UN/ECE, EC, Forest Intensive Monitoring Co-ordinating Institute, 177 pp.
- Draaijers, G.P.J., Bleeker, A., van der Veen, D., Erisman, J.W., Mols, J., Fonteijn, P., Geusebroek, M., 2001. Field intercomparison of throughfall, stemflow and precipitation measurements performed within the framework of the Pan European Intensive Monitoring Program of EU/ICP forests. TNO report R 2001/140. TNO-MEP, Apeldoorn.
- Draaijers, G.P.J., Erisman, J.W., 1995. A canopy budget model to assess atmospheric deposition from throughfall measurements. *Water, Air and Soil Pollution* 85, 2253–2258.
- Draaijers, G.P.J., Erisman, J.W., Spranger, T., Wyers, G.P., 1996. The application of throughfall measurements for atmospheric deposition monitoring. *Atmospheric Environment* 30, 3349–3361.
- Emberson, L.D., Wieser, G., Ashmore, M.R., 2000. Modelling stomatal conductance and ozone flux of Norway spruce: comparison with field data. *Environmental Pollution* 109, 393–402.
- Emberson, L.D., Ashmore, M., Simpson, D., Tuovinen, J.-P., Cambridge, H.M., 2001. Modelling and mapping ozone deposition in Europe. *Water, Air and Soil Pollution* 130, 577–582.
- EMEP/CCC, 1996. EMEP Manual for Sampling and Analysis. EMEP/CCC Report 1/95.
- Erisman, J.W., Draaijers, G.P.J., 1995. Atmospheric Deposition in Relation to Acidification and Eutrophication, Studies in Environmental Research, vol. 63. Elsevier, Amsterdam, 405 pp.
- Erisman, J.W., Wyers, G.P., 1993. Continuous measurements of surface exchange of  $\text{SO}_2$  and  $\text{NH}_3$ ; implications for their possible interaction in the deposition process. *Atmospheric Environment* 27A, 1937–1949.
- Erisman, J.W., Draaijers, G., Duyzer, J., Hofschreuder, P., van Leeuwen, N., Römer, F., Ruijgrok, W., Wyers, P., Gallagher, M.,

1997. Particle deposition to forests—summary of results and application. *Atmospheric Environment* 31, 321–332.
- Erisman, J.W., Mennen, M.G., Fowler, D., Flechard, C.R., Spindler, G., Grüner, A., Duyzer, J.H., Ruigrok, W., Wyers, G.P., 1998a. Deposition monitoring in Europe. *Environmental Monitoring and Assessment* 53, 279–295.
- Erisman, J.W., Bleeker, A., van Jaarsveld, J.A., 1998b. Evaluation of ammonia emission abatement on the basis of measurements and model calculations. *Environmental Pollution* 102, 269–274.
- Erisman, J.W., Hensen, A., Fowler, D., Flechard, C., Grüner, A., Spindler, G., Duyzer, J., Weststrate, H., Römer, F., Vonk, A., Jaarsveld, H., 2001. Dry deposition monitoring in Europe. *Water, Air and Soil Pollution* 1, 17–27.
- Erisman, J.W., Mols, H., Fonteijn, P., Geusebroek, M., Draaijers, G., Bleeker, A., van der Veen, D., 2003. Field intercomparison of precipitation measurements performed within the framework of the Pan European Intensive Monitoring Program of EU/ICP Forest. *Environmental Pollution* 125, 139–155.
- Flechard, C.R., Fowler, D., Sutton, M.A., Cape, J.N., 2000. A dynamic chemical model of bi-directional ammonia exchange between semi-natural vegetation and the atmosphere. *Quarterly Journal of the Royal Meteorological Society* 125, 2611–2641.
- Fowler, D., Erisman, J.W., 2003. Biosphere/atmosphere exchange of pollutants; Overview of subproject BIATEX-2. In: Midgley, P.M., et al. (Ed.), *Towards Cleaner Air For Europe—Science, Tools And Applications*. Margraf, Weikersheim, pp. 35–58.
- Fowler, D., Flechard, C., Cape, J.N., Storeton-West, R.L., Coyle, M., 2001. Measurements of ozone deposition to vegetation quantifying the flux, the stomatal and non-stomatal components. *Water, Air and Soil Pollution* 130, 63–74.
- Grünhage, L., Haenel, H.-D., 1997. PLATIN (PLant-ATmosphere Interaction) I: a model of plant-atmosphere interaction for estimating absorbed doses of gaseous air pollutants. *Environmental Pollution* 98, 37–50.
- Grünhage, L., Haenel, H.-D., 2000. WINDEP—Worksheet-INtegrated Deposition Estimation Programme. In: KRdL—Kommission Reinhaltung der Luft im VDI und DIN (Hrsg.): *Troposphärisches Ozon. Eine kritische Bestandsaufnahme über Ursache, Wirkung und Abhilfemaßnahmen*. Schriftenreihe der KRdL, Düsseldorf, pp. 157–173.
- Grünhage, L., Haenel, H.-D., 2001. Spatial variability of meteorological parameters, driving forces and fluxes. In: Midgley, P.M., Reuther, M., Williams, M. (Eds.), *Proceedings from the EURO-TRAC-2 Symposium, 2000*. Springer, Berlin, CD-ROM.
- Grünhage, L., Dämmgen, U., Haenel, H.-D., Jäger, H.-J., 1998. Response of a grassland ecosystem to air pollutants. VI. The chemical climate: concentrations and potential flux densities of relevant criteria pollutants. *Environmental Pollution* 101, 215–220.
- Grünhage, L., Jäger, H.-J., Haenel, H.-D., Löpmeier, F.-J., Hanewald, K., 1999. The European critical levels for ozone: improving their usage. *Environmental Pollution* 105, 163–173.
- Grünhage, L., Dämmgen, U., Erisman, J.W., Lüttich, M., Hanewald, K., Jäger, H.-J., Freitag, K., Baltrusch, M., Liebl, K., 2002. Atmospheric nitrogen dynamics in Hesse, Germany: The challenge and its potential solution. *Landbauforschung Völkenrode* 52, 219–228.
- Grünhage, L., Krupa, S.V., Legge, A.H., Jäger, H.-J., 2004. Ambient flux-based critical values of ozone for protecting vegetation: differing special scales and uncertainties in risk assessment. *Atmospheric Environment* 38, 2433–2437.
- Haenel, H.-D., Grünhage, L., 1999. Footprint analysis: A closed analytical solution based on height-dependent profiles of wind speed and eddy viscosity. *Boundary-Layer Meteorology* 93, 395–409.
- Haenel, H.-D., Grünhage, L., 2001. Reply to the Comment on ‘Footprint analysis: a closed analytical solution based on height-dependent profiles of wind speed and eddy viscosity’, by T.W. Horst. *Boundary-Layer Meteorology* 101, 449–458.
- Hole, L.R., Semb, A., Tørseth, K., 2004. Ozone deposition to a temperate coniferous forest in Norway; gradient method measurements and comparison with the EMEP deposition module. *Atmospheric Environment* 38 (15), 2217–2223.
- Horst, T.W., 1997. The footprint for estimation of atmosphere-surface exchange fluxes by eddy-correlation and profile techniques. In: *Preprints of the Special Symposium on Boundary Layers and Turbulence (Land Surface)*, 77th AMS Annual Meeting, 2–7 February 1997, Long Beach, CA. American Meteorological Society, Boston, MA.
- Horst, T.W., 1999. The footprint for estimation of atmosphere-surface exchange fluxes by profile techniques. *Boundary-Layer Meteorology* 90, 171–188.
- Horst, T.W., Weil, J.C., 1992. Footprint estimation for scalar flux measurements in the atmospheric surface layer. *Boundary-Layer Meteorology* 59, 279–296.
- Jung, C.H., Kim, Y.P., Lee, K.W., 2003. A moment model for simulating raindrop scavenging of aerosols. *Journal of Aerosol Science* 34, 1217–1233.
- Kormann, R., Meixner, F.X., 2001. An analytical footprint model for non-neutral stratification. *Boundary-Layer Meteorology* 99, 207–224.
- Loosmore, G.A., Cedervall, R.T., 2004. Precipitation scavenging of atmospheric aerosols for emergency response applications: Testing an updated model with new real-time data. *Atmospheric Environment* 38, 993–1003.
- Ludwig, J., Meixner, F.X., Vogel, B., Förstner, J., 2001. Soil-air exchange of nitric oxide: An overview of processes, environmental factors, and modelling studies. *Biogeochemistry* 52, 225–257.
- Nemitz, E., Milford, C., Sutton, M.A., 2001. A two-layer canopy compensation point model for describing bi-directional biosphere/atmosphere exchange of ammonia. *Quarterly Journal of the Royal Meteorological Society* 127, 815–833.
- Pilegaard, K., 2001. Air-soil exchange of NO, NO<sub>2</sub> and O<sub>3</sub> in forests. *Water, Air and Soil Pollution* 1, 79–88.
- Schaug, J., Iversen, T., Pedersen, U., 1993. Comparison of measurements and model results for airborne sulfur and nitrogen components with Kriging. *Atmospheric Environment* 27A, 831–844.
- Schmid, H.P., 1994. Source areas for scalars and scalar fluxes. *Boundary-Layer Meteorology* 67, 293–318.
- Simpson, D., Tuovinen, J.-P., Emberson, L.D., Ashmore, M.R., 2001. Characteristics of an ozone deposition module. *Water, Air and Soil Pollution: Focus* 1, 253–262.
- Smith, R.I., Fowler, D., Sutton, M.A., Flechard, C., Coyle, M., 2000. Regional estimation of pollutant gas dry deposition in the UK: model description, sensitivity analyses and outputs. *Atmospheric Environment* 34, 3757–3777.
- Sutton, M.A., Nemitz, E., Fowler, D., Wyers, G.P., Otjes, R.P., San José, R., Moreno, J., Schjørring, J.K., Husted, S., Meixner, F.X., Ammann, C., Neftel, A., Gut, A., 1996. Exchange of atmospheric ammonia with European ecosystems (EXAMINE). Final Report to the European Commission, vol. 1 (EV5V-CT94-0426). Institute of Terrestrial Ecology, Edinburgh, 266 pp.
- Sutton, M.A., Burkhardt, J.K., Guerin, D., Nemitz, E., Fowler, D., 1998. Development of resistance models to describe measurement of bi-directional ammonia surface-atmosphere exchange. *Atmospheric Environment* 32, 473–480.
- Sutton, M.A., Milford, C., Nemitz, E., Theobald, M.R., Hill, P.W., Fowler, D., Schjørring, J.K., Mattson, M.E., Nielsen, K.H., Husted, S., Erisman, J.W., Otjes, R., Hensen, A., Mosquera, J., Cellier, P., Loubet, B., David, M., Genermont, S., Neftel, A., Blatter, A., Herrmann, B., Jones, S.K., Horvath, L., Fuhrer, E.C., Mantzanas, K., Koukoura, Z., Gallagher, M., Williams, P., Flynn, M., Riedo, M., 2001. Biosphere-atmosphere interactions of

- ammonia with grasslands: Experimental strategy and results from a new European initiative. *Plant and Soil* 228, 131–145.
- Tarrason, L., Schaug, J., 2000. Transboundary acidification and eutrophication in Europe. EMEP summary report CCC and MSCW (EMEP Report 1/2000). Oslo.
- Tuovinen, J.-P., 2000. Assessing vegetation exposure to ozone: properties of the AOT40 index and modifications by deposition modelling. *Environmental Pollution* 109, 361–372.
- Tuovinen, J.-P., Simpson, D., Mikkelsen, T.N., Emberson, L.D., Ashmore, M.R., Aurela, M., Cambridge, H.M., Hovmand, M.F., Jensen, N.O., Laurila, T., Pilegaard, K., Ro-Poulsen, H., 2001. Comparison of measured and modelled ozone deposition to forests in Northern Europe. *Water, Air and Soil Pollution: Focus* 1, 263–274.
- Tuovinen, J.-P., Ashmore, M.R., Emberson, L.D., Simpson, D., 2004. Testing and improving the EMEP deposition module. *Atmospheric Environment* 38, 2373–2385.
- Ulrich, B., 1983. Interaction of forest canopies with atmospheric constituents: SO<sub>2</sub>, alkali and earth alkali cations and chloride. In: Ulrich, B., Pankrath, J. (Eds.), *Effects of Accumulation of Air Pollutants in Forest Ecosystems*, pp. 33–45.
- Van Leeuwen, E.P., Draaijers, G.P.J., Potma, C., van Pul, W.A.J., Erisman, J.W., 1996. The compilation of measurement based European wet deposition maps of acidifying components and base cations. *Water, Air and Soil Pollution* 85, 2173–2178.
- Wesely, M.L., Hicks, B.B., 2000. A review of knowledge on dry deposition. *Atmospheric Environment* 34, 2261–2282.
- Zimmermann, W., 1825. Beiträge zur näheren Kenntniss der wasserigen Meteore. *Archiv für die gesammte Naturlehre* 1, 257–293.