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Dry deposition of particles

Implications and recommendations for mapping of deposition over Europe*

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ABSTRACT

This paper assesses current knowledge about the dry deposition of particles with a focus on implications for Europe. General aspects of sources, physical characteristics and concentration data of particles in the atmosphere are considered. Current modelling approaches for calculating the dry deposition of small and coarse particles are reviewed. Shortcomings and uncertainties of these models are presented. Measurements of small- and large-particle deposition with various techniques are summarized together with their limitations. Finally, implications of scale aspects of particle dry deposition are treated with an emphasis on Europe. The overall conclusion is that knowledge on particle dry deposition is still insufficient, both experimentally and theoretically, undermining reliable estimates of deposition fluxes over Europe.

1. Introduction

Together with wet deposition, particle dry deposition is responsible for delivering to ecosystems atmospheric loads of compounds such as SO_4^{2-}, NO_3^- and NH_4^+, base cations (such as Na^+, K^+, Ca^{2+} and Mg^{2+}) and heavy metals (e.g., Pb, Cd and Zn). Deposition of particles containing SO_4^{2-}, NO_3^- and NH_4^+ contributes to potential acidification and eutrophication of ecosystems. Base cation deposition can be important for nutrient cycling in soils and ecosystems and it can also neutralise acid input. Heavy metals are known for their possible toxic effects. Here, we will focus on the dry deposition of both the acidifying particles and base cations.

Despite the importance of this topic, dry deposition of particles has received far less attention than the deposition of acidic gases. This is partly because of results of wind tunnel studies suggesting that the deposition velocity may be very small for submicron particles. Also, the complexity of deposition processes of particles implies that it is difficult to perform field measurements appropriately.

Current knowledge is insufficient to give an adequate assessment of the dry deposition of particulate sulphur and nitrogen over Europe. The same holds for the fluxes of alkaline particles containing base cations. However, there is a need for a proper quantification of both types of fluxes to evaluate critical loads of acidic deposition and abatement strategies of atmospheric pollution.

In this paper, we will provide an appraisal of current knowledge about particle dry deposition and of possibilities to infer fluxes from meteorological and concentration data, topography and land-use characteristics. Scale aspects and uncertainties of flux determinations are also discussed. An emphasis is placed on implications for Europe.

This appraisal is structured as follows. First, we

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* This paper is based on a background document which was prepared for discussions to assess the current state of knowledge at a workshop on deposition organized by the Nordic Council of Ministers and EMEP (Gothenburg, Sweden, 3–6 November 1992).

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present information on the nature of particles in the atmosphere, giving data on concentrations and emission sources as well as physical characteristics of the particles. Then, we review deposition models and summarize experimental data from the literature. Finally, we synthesize this information to reach conclusions on current knowledge of particle dry deposition and on ways to derive deposition fluxes of particles.

2. Particles in the atmosphere

2.1. Sources

The contribution of anthropogenic sources to particulate SO_4^{2-}, NO_3^- and NH_4^+ in the atmosphere is mainly the result of emissions of the gaseous precursors SO_2, NO_x and NH_3. In the atmosphere both SO_2 and NO_x are subject to chemical transformation processes by oxidants such as H_2O_2, O_3 and OH radicals, which lead to the formation of H_2SO_4-containing aerosol and gaseous HNO_3. These two products may then react further under typical European conditions with NH_3, so that both SO_4^{2-} and NO_3^- are in the aerosol phase. This chemical transformation process is generally referred to as "gas-to-particle" conversion giving "secondary aerosol". Gas-to-particle conversion rates are low for SO_2 (of the order of 1–2% per hour) but can be 10–20 times higher for NO_x. The chemical transformation of gases into particles involves many factors and issues on which details can be found elsewhere (Finlayson-Pitts and Pitts, 1986; Seinfeld, 1986; Twomey, 1977).

Another route to form particles is via non-precipitating clouds. On average, 90% of the cloud droplets existing at any instant of time will evaporate (Junge, 1963). Therefore, chemistry taking place in cloud water involving conversion of SO_2, NO_x, NH_3 and other compounds can produce aerosols that are left after droplet evaporation. For SO_2 and NH_3 this droplet-to-particle conversion appears to be an efficient route to produce $(NH_4)_2SO_4$ aerosol. Secondary aerosols comprising $(NH_4)_2SO_4$ are stable and will remain in the atmosphere until removed by wet or dry deposition. Aerosols containing NH_4NO_3, however, are unstable and may dissociate again, for instance under atmospheric conditions with low NH_3 concentrations.

Natural sources can emit aerosols as well as gaseous precursors. For example, sulphur compounds are emitted in both phases from volcanoes and the oceans. Wind erosion of soil is a well known source of large particles, which can be neutralizing agents for acidic compounds. Source strengths have been quantified on a global scale in the literature (e.g., in Lee et al., 1986, and in Schwartz and Slinn, 1992), especially for sulphur, but ranges and uncertainties are considerable.

2.2. Physical characteristics

Particles in the atmosphere cover a wide size range, from <0.01 μm (Aitken particles) to >100 μm for soil particles. The peak of aerosol mass of anthropogenic origin is normally in the 0.1–1 μm range, due to the underlying transformation processes of gaseous precursors. The mass distribution function of anthropogenic SO_4^{2-} aerosol exhibits a log-normal distribution characterized by a mass median diameter (MMD) of 0.48 ± 0.10 μm and geometric standard deviation σ_g of 2.00 ± 0.29 (Whitby, 1978). Milford and Davidson (1987) reviewed 42 observations of SO_4^{2-} size distributions from continental locations (urban and nonurban). As an average MMD they report 0.52 ± 0.23 μm. The same paper shows that 7 size distributions from marine areas have an MMD of 2.3 ± 1.8 μm. For NH_4^+, Whitby gives an MMD of 0.38 ± 0.08 μm and $\sigma_g = 1.95 \pm 0.38$.

Size distributions may show a bimodal or even multi-modal shape. Most SO_4^{2-} in sea spray will reside on large particles, with MMD ≈ 5 μm and $\sigma_g \approx 2.7$ (Whitby, 1978). Elements such as Na, K and Ca also reside on large particles with similar values for MMD and σ_g (Rahn, 1975). NO_3^- of anthropogenic origin may be formed by reaction of gaseous HNO_3 with NH_3. But HNO_3 can also react with alkaline species residing on large particles which results in a size distribution for NO_3^- that contains both submicron and supermicron particles.

2.3. Concentration data

SO_4^{2-}, NO_3^- and NH_4^+ are formed during long-range transport of gaseous precursors. The residence time of these particles is about a week which enables travel over large distances. As a result concentration patterns tend to show more large scale features than local variations. Concentration patterns of these compounds in Europe

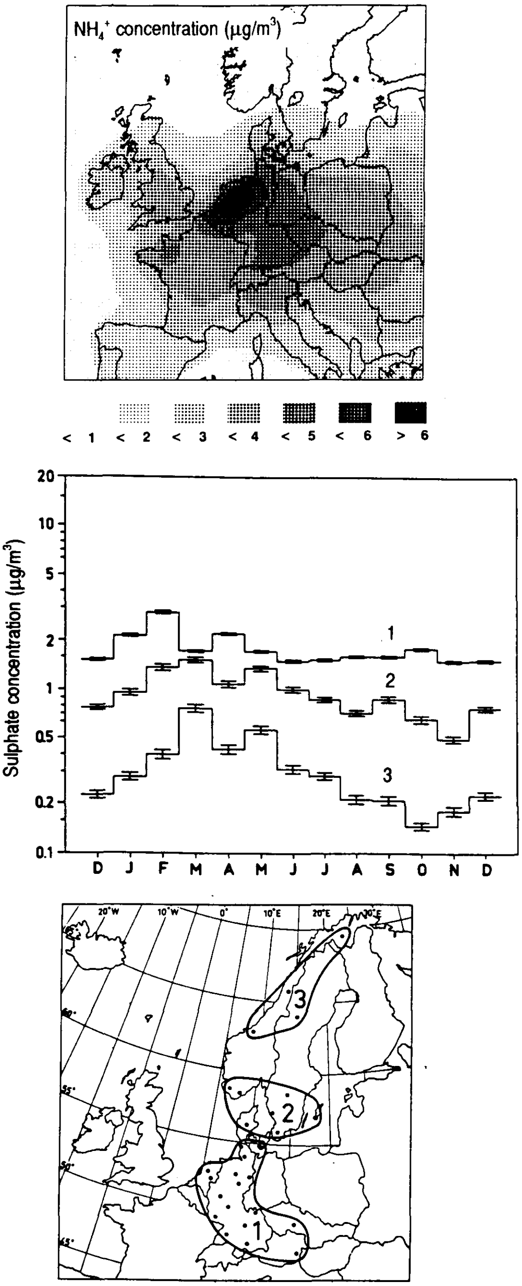

Fig. 1. Concentration field of NH_4^+ over Europe as calculated by the TREND model (Asman and Van Jaarsveld, 1991). Temporal variations in monthly mean SO_4^{2-} concentrations of three European regions according to monitoring results of EMEP (the map in the bottom panel identifies the EMEP-stations in these regions).

Table 1. *Correlation distances for sulphate, ammonium, chloride and nitrate aerosol found in the Netherlands*

SO_4^{2-}	120–140 km	NO_3^-	60 km	(entire year)
NH_4^+	50 km	NO_3^-	>180 km	(winter)
Cl^-	30 km	NO_3^-	30 km	(summer)

can be obtained from results of the EMEP* network (Iversen et al., 1991). As an illustration of the NH_4^+ concentration field over Europe, results obtained with the TREND model (Asman and Van Jaarsveld, 1991) are given in Fig. 1a. Highest concentrations are found over Western and Central Europe. To indicate temporal variations average monthly SO_4^{2-} concentrations are depicted in Fig. 1b for three European regions. SO_4^{2-} decreases in concentration from Central Europe going northwards. A clear seasonal cycle is apparent in northern Scandinavia, but less pronounced close to source regions of SO_2.

Results of a measuring network in the Netherlands (Janssen et al., 1989) provide more information on the interpolation error when deriving a concentration field from measurements. These errors depend on network density and the spatial scale of aerosol concentration patterns. The spatial scale of concentration patterns can be estimated using a correlation distance which is the distance between two stations x and y for which 50% of the variance in concentrations at station x can be explained by the measurements at station y (Table 1). Concentrations of SO_4^{2-} (year-round), as well as NO_3^- (during wintertime) measured at one station appear to be representative of a large area (about the size of a grid cell in the model operated for EMEP, Iversen et al., 1991). NH_4^+, Cl^- and NO_3^- concentrations during the summer season show local influences. The minimum error introduced by interpolation of concentrations from network scale to a finer resolution is on the order of 30–40% (Fig. 2). Interpolation errors are seasonally dependent for NO_3^-.

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* A network of monitoring stations of air pollutants operated by national institutes and authorities under the Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutant in Europe (EMEP) of the United Nations Economic Commission for Europe (UN ECE).

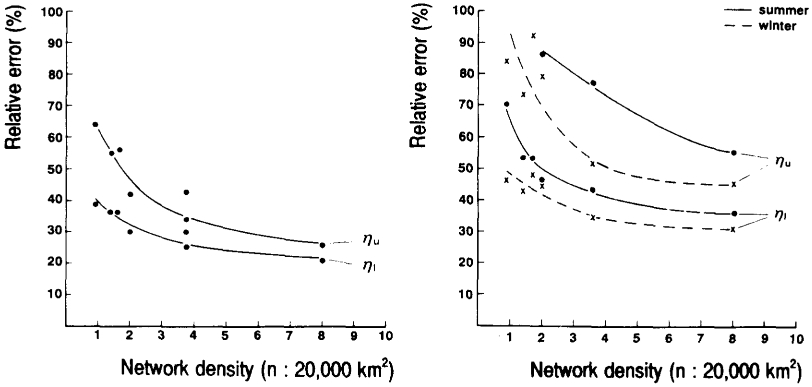

Fig. 2. The interpolation error of concentration fields as a function of network density as derived by Janssen et al. (1991). The left panel shows the error for SO_4^{2-} and the right panel for NO_3^-; η_l indicates the lower limit and η_u the upper limit of the interpolation error.

Concentration data for Na^+, K^+, Mg^{2+} and Ca^{2+} are of particular interest for the interpretation of throughfall data and the evaluation of critical loads (Hettelingh et al., 1991). Despite this importance, systematic monitoring is lacking in Europe because measurements of these elements are difficult to perform and were considered of little interest until recently. Among the complicating factors in measuring are the representativity in space and the collection efficiency of sampling. Hettelingh et al. (1991) calculated concentration fields of these compounds for a European critical load assessment. Their method consisted of converting precipitation concentrations into ambient concentrations by using scavenging ratios. However, the results of this method are highly uncertain, because rain events occur only a small fraction of time when airborne concentrations may not be representative of yearly averages. Furthermore, the use of scavenging ratios has been frequently criticized on physical grounds (Barrie, 1992).

3. Deposition models

It is convenient to consider dry deposition of particles as occurring in three steps. In the first step, particles are transported from the free atmosphere down to the viscous sublayer that envelops all surfaces. Second, the particles are transported across the viscous sublayer by Brownian diffusion, phoretic effects, interception, inertial forces such as impaction, and sedimentation. Finally, the particles interact with the surface: they may adhere or they may bounce off. These processes, or at least some of them, are incorporated into model formulations.

The dry deposition velocity v_d is often used to

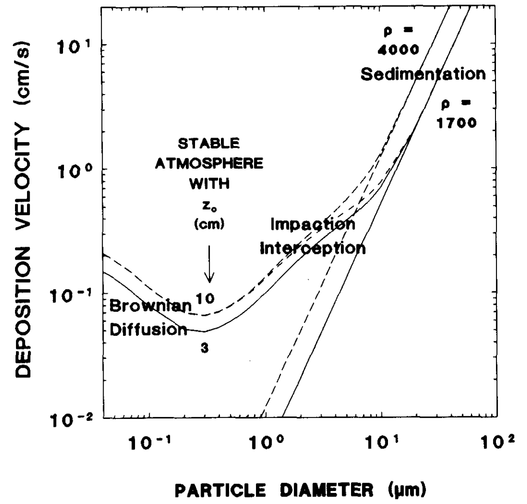

Fig. 3. Typical curves for the deposition velocity of particles as a function of size in a stable atmosphere for two different roughness lengths (z_0 in cm) and particle densities (ρ in kg m^{-3}). The settling velocity of particles is indicated also. The curves were calculated with $u = 5$ m/s using the model described by Sehmel and Hodgson (1980).

Table 2. *Characteristic features of some models describing the dry deposition of particles to vegetation (grass and forests) and water surfaces*

				Processes included in the model								
	Surface	Type	Diameter range	Turbulent transport	Sedimentation	Impaction	Interception	Brownian diffusion	Rebound	Hygroscopic growth	Capture by waves	Shown in Fig. 4
Sehmel & Hodgson (1980)	any	A/I	10^{-3}–10^2	+	§	§	§	+	–	–		yes
Haynie (1986)	any	A	10^{-3}–10^2	+ *	+	+	–	+	–	–		
Schack et al. (1985)	water/vegetation	A	10^{-3}–10^2	+ *	+	§	§	§	–	–		
Ibrahim et al. (1983)	snow/forest	A	10^{-3}–10^2	+ *	+/–	+	+	+	–	–		
Wiman and Ågren (1985)	forest	N	10^{-3}–10^2	+	+	+	–	+	+/–	–		yes
Peters and Eiden (1992)	forest	A/I	10^{-3}–10^2	+ *	+	+	+	+	–	–		
Davidson et al. (1982)	grass	N	10^{-3}–10^2	+ *	+	+	+	+	+/–	–		yes
Legg & Price (1980)	vegetation	A/I	10–10^2	+ *	–	–	–	–	–	–		yes
Bache (1979)	vegetation	N	10–10^2	+ *	+	+	–	–	–	–		yes
Slinn (1982)	vegetation	A	10^{-3}–10^2	+ *	+	+	+	+	+	–		yes
Slinn & Slinn (1980)	water	A	10^{-3}–10^2	+ *	+	+	+	+	–	+	–	
Williams (1982)	water	A	10^{-3}–10^2	+ *	+	+	+	+	–	+	+	yes
Hummelshoj et al. (1992)	water	A	10^{-3}–10^2	+ *	+	+	+	+	–	+	+	

+ : mechanism included.
– : mechanism not included in the model.
§: mechanism included by empirical data-fit.
+/– : considered, but not included.

*: only for neutral conditions.
A: analytical model.
N: numerical model.
A/I: analytical with iterative solving for some parts.

quantify the deposition process. The deposition velocity v_d is defined as the flux divided by the airborne concentration at a certain reference height. The deposition velocity is highly dependent on particle size with different removal processes at work in different size regions.

Several models are available describing the dry deposition velocity of particles as a function of particle size, based on mathematical relations describing each removal process. We will refer to these types of models as *process oriented models*. Another way of modelling particle deposition is by using a simple surface resistance approach without making distinction for particle size. This method is commonly employed for dry deposition of trace gases; here, it will be referred to as *bulk resistance oriented models*.

3.1. Process oriented models

A typical example of the deposition velocity as a function of particle size is given in Fig. 3. Responsible collecting mechanisms are identified for three size regions. Preferably, all these collecting mechanisms should be included in process oriented deposition models. Several such models are now available for calculating aerosol deposition to various surfaces under different conditions. Table 2 summarizes the main features of a number of these process oriented models. Phoretic effects (electrophoresis, thermophoresis, diffusiophoresis) are not treated in these deposition models. However, Slinn (1982) suggests that electrophoresis (deposition due to electrical charges of particles) could be of special interest for small particles. Hygroscopic growth of particles is included in three models only. Such growth of particles is possible at high relative humidities (rh > 80%; Fitzgerald, 1975) and will result in a more efficient removal of particles by impaction and interception.

A major shortcoming of many of the models gathered in Table 2 is that they deal only with neutral atmospheric conditions. This may overestimate v_d under stable conditions and underestimate v_d for unstable conditions, possibly by a factor of 2–3 or more (Sievering, 1989) if deposition is controlled by atmospheric transport. Many of these models were validated only against wind tunnel measurements, which may not be representative of field situations.

Assessments of model uncertainties are usually not reported by the authors. Because these

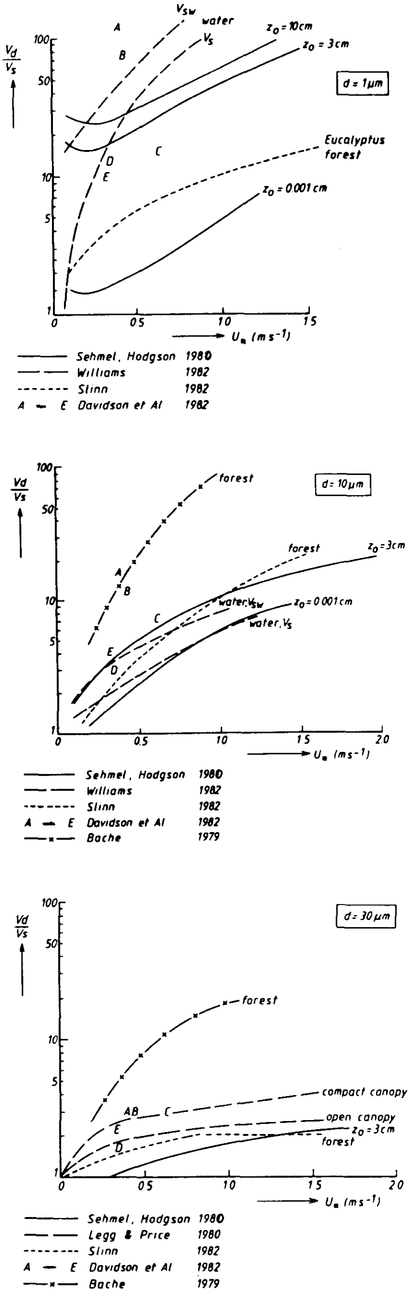

Fig. 4. Comparison of the dependence of v_d on the friction velocity u_* as presented by Van Aalst (1986) for 6 models for various surfaces and conditions (v_d is expressed here relative to the particle settling velocity, v_g). Some features of the models displayed are presented in Table 2.

models incorporate very different formulations and assumptions, it is difficult to compare model results quantitatively. Nevertheless, an attempt at such a comparison has been presented by Van Aalst (1986). Fig. 4 summarizes the results for 1, 10 and 30 μm particles. The model of Williams (1982) for a smooth water surface gives deposition velocities 10 to 50 times as high for 1 μm particles as the model of Sehmel and Hodgson (1980) for controlled roughness conditions. The model of Slinn (1982) for a forest canopy results in lower v_d values than predicted by Davidson et al. (1982) for grass (1 μm particles) although a forest canopy is characterized by a considerably larger roughness length. Differences in v_d between the various models tend to be smaller for larger particles, except for forest applications. The rebound of particles is visible for 30 μm particles and large values of u_* in the model of Slinn. Recent data suggest that particle bounce-off and resuspension are major factors influencing deposition from ambient surfaces (Wu et al., 1992).

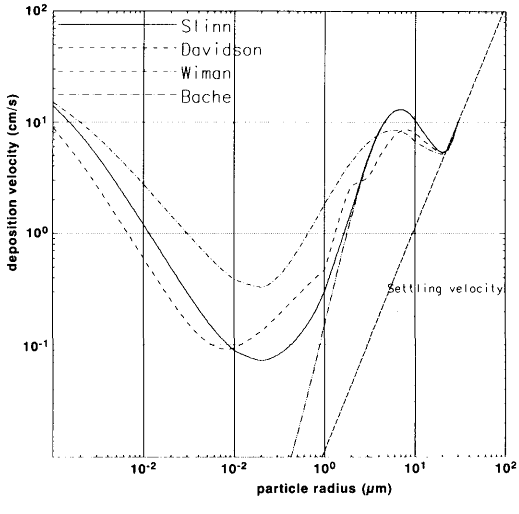

Fig. 5. Variation in predicted deposition velocities due to different mathematical formulations of deposition mechanisms when applied within the model framework of Slinn (1982) for a *Eucalyptus* forest. Formulations of Bache (1979), Davidson et al. (1982), Wiman and Ågren (1985) and Slinn (1982) were used for Brownian diffusion, interception and impaction, respectively. Rebound of particles was treated in the same way in each calculation according to the original formulation of Slinn (1982). Some parameter values used are $u = 5$ m/s ($z = 37$ m), $z_0 = 1$ m and canopy height $h = 27$ m.

The mathematical expressions used in models to describe deposition mechanisms as diffusion, impaction and interception vary, all of which are based on fundamental aerosol research (be it experimental or theoretical). For Fig. 5, some of these expressions used in other models have been implemented in Slinn's model for calculating the deposition to forested areas. Rebound of particles is included in the calculation and treated in the same way for all approaches. As can be seen (Fig. 5) there is a considerable range in v_d, especially for 0.1–1 μm particles, due to the description of mechanisms only. Wiman and Ågren's (1985) formulation gives values for v_d about a factor 5 larger than those of Slinn.

Yet another way to identify uncertainty and variability of v_d is to use appropriate frequency distributions for model input parameters. Various values drawn randomly from these frequency distributions can then be used to run the model. If this procedure is repeated many times, one can construct a probability distribution of v_d. Results for Slinn's model are shown in Fig. 6 from two studies. Fig. 6a gives results of a study (Ruijgrok, 1992) in which variable input values for 9 different parameters were used to determine a probability distribution function of v_d. These parameters concerned the size of small and large collectors in the vegetation, the fraction of momentum collected by small elements, the shape of the wind profile in the canopy, the ratio of viscous and total drag in the canopy, particle density, tree height, roughness length and wind speed. The largest uncertainty is located in the 0.1–1 μm size interval: values can vary over 2 to 3 orders of magnitude. The main factors as determined in this study for the uncertainty in v_d are the uncertainty and variability in the size of particle-collecting elements within the canopy, roughness length and wind speed.

Gould and Davidson (1992) addressed the uncertainty in v_d as calculated by the model of Slinn as well. They determined the influence of uncertainty in the size of small and large collectors, roughness length, the canopy wind profile and wind speed. The uncertainty in v_d was assessed in 5 stages. First variation in the size of small collectors was included only. The second step included variation in small and large collectors. In the next three steps variation in roughness length, the canopy wind profile and wind speed were added, respectively. Results are given in Fig. 6b for the

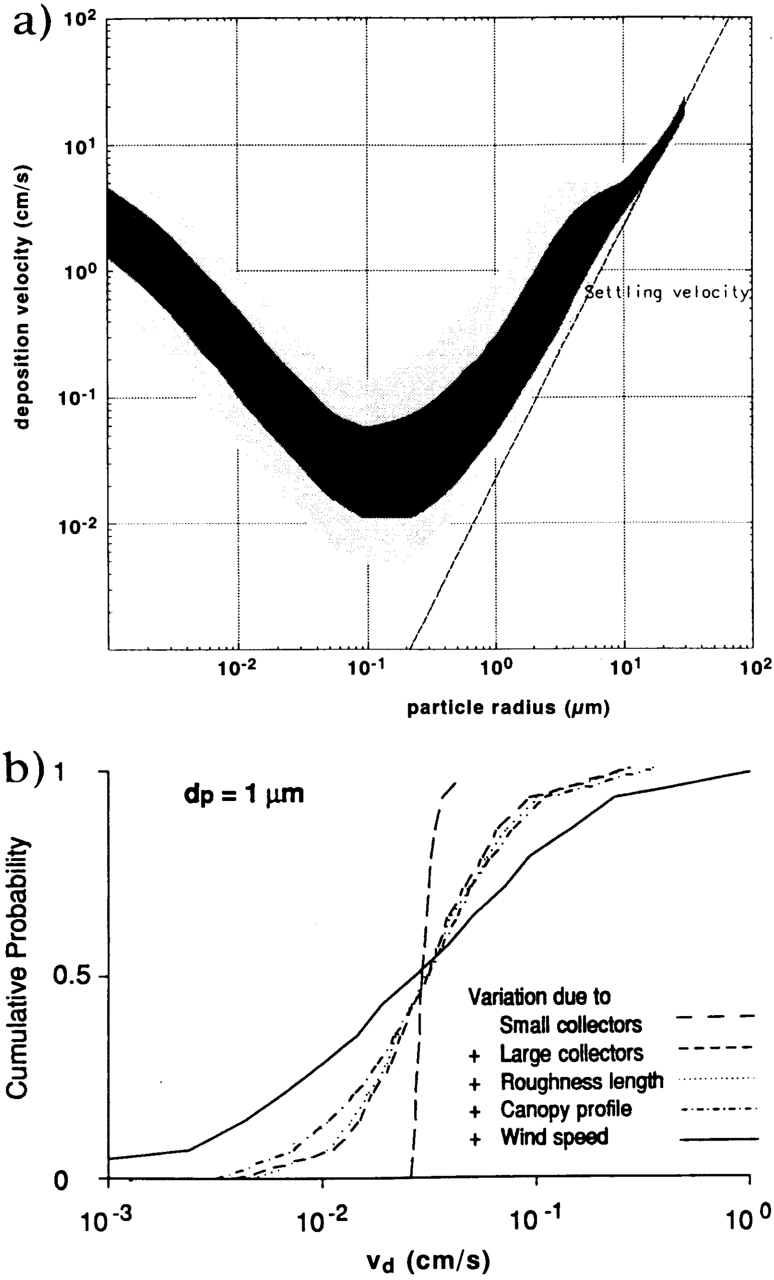

Fig. 6. Uncertainty in v_d due to uncertain and variable input parameters of the deposition model by Slinn (1982). (a) The uncertainty in v_d depending on particle size covering 60% (light grey) and 90% (dark grey) of variation around the median v_d predicted due to variatiability in 9 input parameters (Ruijgrok, 1992). (b) The evolution of the cumulative probability function of v_d for 1 μm particles due to the stepwise variation of 5 input parameters (Gould and Davidson, 1992). The median v_d values in (a) and (b) differ due to different median values used for input parameters.

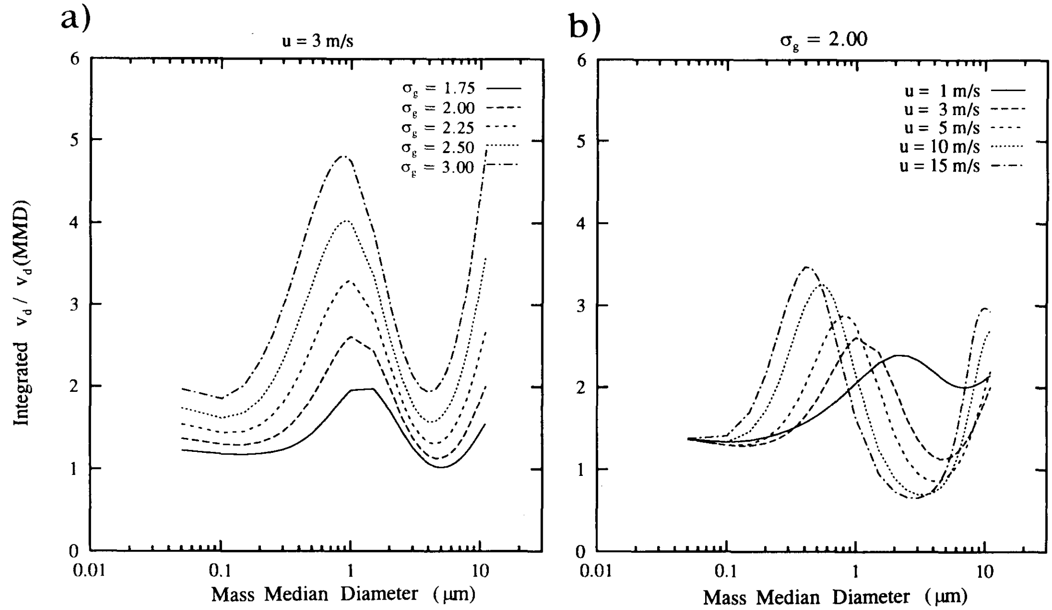

Fig. 7. Integrated values for v_d as a function of mass median diameter depending on geometric standard deviation (left panel) and wind speed (right panel). Integrated values of v_d are expressed relative to the v_d for a particle with a diameter equal to the mass median diameter. The increase in this ratio for MMD values larger than 5 μm is due to the incorporation of particle rebound in the model calculations. Fig. 7a was calculated with $u = 3$ m/s ($z = 37$ m) and Fig. 7b with $\sigma_g = 2.0$. Calculations are with Slinn's model for forest conditions ($z_o = 1$ m, canopy height $h = 27$ m).

evolution of the cumulative function of v_d for 1 μm particles due to this stepwise introduction of variable input parameters. The median v_d values of Figs. 6a, b differ due to different median values used for input parameters. However, both studies identify similar parameters as the main factors determining the uncertainty in v_d.

It is of interest to compare values for v_d computed for an entire particle distribution with those computed for the single value of MMD for this distribution. Differences between these results show the importance of integrating a model over all particle sizes rather than simply using the MMD. Integrated values of v_d are shown in Fig. 7

Table 3. *Values for r_b (s cm^{-1}) in the resistance approach of Voldner et al. (1986) for particulate SO_4^{2-}; r_b for NO_3^- is suggested to be taken as half the value for SO_4^{2-}*

	Winter		Spring		Summer		Fall	
Surface	day	night	day	night	day	night	day	night
coniferous forest	1.4	3	1.4	3	1.4	3	1.4	3
deciduous forest	10	20	3	6	1	2	2	4
cultivated land	10	25	5	10	3	6	5	10
grass	10	25	4	6	3	6	3	6
urban	20	40	6	12	2	4	4	8
swamp	10	25	3	4	2	4	2	4
open water	0	0	0	0	0	0	0	0
snow/ice	10	25	—	—	—	—	—	—

depending on mass median diameter, geometric standard deviation of the size distribution and wind speed. For particles distributed with a mass median diameter of around 0.5 μm and σ_g between 1.75 and 3.00, v_d integrated over the entire size range is about 3–6 times higher than the value of v_d corresponding to a particle with 0.5 μm diameter.

3.2. Bulk resistance models

Instead of the detailed process oriented models, several methods describe deposition in terms of resistances or deposition velocities which are thought to be applicable over the entire size range of depositing particles. The name *bulk resistance models* is used here to indicate that size dependency is not explicit but accounted for in the resistance value by integration.

Voldner et al. (1986) have proposed to parameterize v_d for anthropogenic SO_4^{2-} and NO_3^- by $v_d = (r_a + r_b)^{-1}$, in which r_a represents the well known aerodynamic resistance and r_b the resistance of the viscous sublayer surrounding collecting surfaces. Their "best" values for r_b are given in Table 3. These values are based on either very small data sets or on analogies, and hence there is considerable uncertainty in the distinction between day and night values and between surface types.

Hicks et al. (1987) propose a resistance scheme which parameterizes v_d as

$$v_d = (r_a + r_{cp} + r_a r_{cp} v_g)^{-1} + v_g,$$

where v_g stands for particle settling velocity and r_{cp} is a net canopy resistance term to particle deposition incorporating an equivalent of r_b. However, Hicks et al. conclude that present knowledge is so poor that such a scheme cannot be handled properly. Therefore, as an interim measure, they adopted a purely empirical relationship according to work by Wesely et al. (1983). This relationship, which is based on a limited set of conditions, is $v_d = 0.003\sigma_\Theta u$, in which σ_Θ is the standard deviation of wind direction and u is the wind speed.

Wesely et al. (1985) suggested also another parameterization for v_d based on flux measurements of particulate SO_4^{2-} over grassland. First, they removed the influence of aerodynamic resistance on v_d to get a surface deposition velocity v_{ds}:

$$v_{ds} = (v_d^{-1} - r_a)^{-1}.$$

This surface deposition velocity is dependent on atmospheric stability and can be described as a function of the friction velocity u_* and the Monin-Obukhov length, L:

$$v_{ds} = 0.002u_*,$$

$$L > 0 \text{ (stable conditions)},$$

$$v_{ds} = 0.002u_*[1 + (-300/L)^{2/3}],$$

$$L < 0 \text{ (unstable conditions)}.$$

This parameterization was used to calculate v_d values for the US, based on land use characteristics (Sheih et al., 1986), yielding an average of around 0.4 cm/s. In an adjusted form it was also used to derive fluxes of particulate sulphur and nitrogen for 5x5 km^2 grid cells in the Netherlands using measured wind and concentration fields (Erisman, 1993). As a yearly averaged value for v_d Erisman obtained 0.5 cm/s over the Netherlands.

On the basis of his process oriented model, Slinn (1982) gave an "empirical" relationship that might be used to derive values for v_d. However, although simplified, his approach still requires information which is too detailed to apply for generalization of particle deposition fluxes over Europe.

In the EMEP model, particle deposition is accounted for by a uniform v_d, independent of time, surface and atmospheric characteristics. For calculations of NH_4^+ deposition with the TREND model, a constant value of the surface resistance was used ($r_c = 600$ s/m, giving an effective $v_d = 1.4$ mm/s), allowing dependency of v_d on atmospheric stability, but still neglecting surface characteristics. Moreover, the value chosen for r_c is quite arbitrary.

4. Deposition measurements

Nicholson (1988) and Davidson and Wu (1990) gave the most recent reviews of particle deposition measurements. A summary of these two reviews is provided in Table 4. Other reviews were given by, e.g., Sehmel (1980) and Voldner et al. (1986). One factor that should be noted for particle deposition measurements is that such experiments are severely hampered by the dependence of v_d on particle size. In most experiments size ranges have not been determined. This makes it difficult to compare values measured with results from process

Table 4. *A summary of deposition velocities (cm s^{-1}) derived from field experiments (the number of experiments is given as n) using different techniques as reviewed by Nicholson (1988) and Davidson and Wu (1990)*

	Technique	Mean	*n*	Range
SO_4^{2-}	micrometeorological	0.55 ± 0.65	20	0.01–2.9
	foliar extraction	0.23 ± 0.24	5	0.05–1.2
	surrogate surfaces	0.26 ± 0.25	26	0.01–0.87
	throughfall	1.0 ± 0.41	9	0.38–2.0
NO_3^-	foliar extraction	0.21 ± 0.11	7	0.13–0.43
	surrogate surfaces	0.50 ± 0.32	3	0.13–0.69
	throughfall	1.24 ± 0.71	9	0.33–2.25
NH_4^+	foliar extraction	0.44	1	—
	surrogate surfaces	0.20 ± 0.20	7	0.08–0.6
	throughfall	1.0 ± 0.31	7	0.5–1.4
Ca	surrogate surfaces	2.4 ± 1.9	11	0.2–7.0
Mg	surrogate surfaces	1.9 ± 3.5	9	0.2–11.0
Na	surrogate surfaces	1.6 ± 2.1	12	0.2–7.4
K	surrogate surfaces	0.9 ± 0.6	8	0.2–1.9

oriented model calculations. These two remarks have to be kept in mind when one evaluates estimates of v_d from deposition measurements or from model calculations.

A wide variation in measured values of v_d exists, ranging from negative values to several cm s^{-1}. These wide variations reflect different experimental conditions as well as measurement error. Values of v_d values derived from throughfall experiments give especially high values. There should be caution in using data from throughfall experiments in order to derive deposition velocities. First, dry deposition fluxes must be corrected for leaching or uptake by the vegetation, which is not straightforward for all compounds in throughfall. Second, the dry deposition fluxes of SO_4^{2-}, NO_3^- and NH_4^+ must be partitioned into fluxes caused by gaseous and particulate deposition, which is difficult for throughfall experiments.

The use of surrogate surfaces may lead to either the overcollection or undercollection of depositing particles. Further, it is questionable to what extent these devices are representative of natural surfaces. When natural vegetation surfaces are used, leaching or material uptake by the inner plant parts can be a serious problem. For the micrometeorological techniques stringent criteria on spatial homogeneity and temporal stationarity apply for the succesful use of these techniques. However, it is often difficult to fulfill all demands in experiments under field conditions. Misrepresentation due to selection of data can also affect final results. Humidity profiles may have considerable biasing effects on eddy correlation measurements. Besides the errors in the techniques themselves, accurate measurements of the size distribution of airborne material was not involved in most deposition measurements summarized in Table 4. Therefore, it cannot be ruled out that the higher deposition velocities were caused by sedimentation effects of large particles.

The error in v_d obtained from experiments and the uncertainty in model results for submicron particles overlap one another in most cases. However, some recent experimental results gathered in Table 4 show values of v_d substantially larger than ~0.1 cm/s, a value typical for the submicron size range wind tunnel results and of many (but not all) models. The larger values are usually obtained above (very) rough surfaces such as forests. Possible explanations for the orders of magnitude difference between field and theoretical values of v_d include the effect of very rough surfaces such as forests, complex flow around a vegetated surface, the effect of high humidity near surfaces (leading to particle growth) and conditions with high wind speeds. The gap between measured values in field experiments and results from model and wind tunnel work is large and requires clarification. There is a clear need for an intercalibration of the various measuring methodologies and for the calibration of current models against reliably collected field data.

5. Scale aspects of deposition

There is an increasing interest in obtaining deposition estimates over Europe to evaluate emission reduction policies, develop new reduction strategies and assess exceedance of critical loads in vulnerable areas. The concept of critical loads requires information on dry deposition of acidifying as well as alkaline compounds, preferably on the regional and local scale. In this section we discuss some of the difficulties of applying deposition estimates on the scales needed for policy development.

5.1. European and regional scale

A major difficulty with the application of models for predicting deposition is their spatial resolution. Estimates of deposition on the European scale have been made with the EMEP model for grids with a 150x150 km^2 size (Iversen et al., 1991; Eliassen and Saltbones, 1983). Similar estimates have been made with a higher resolution using the TREND model (e.g., for NH_3/NH_4^+, Asman and Van Jaarsveld, 1991). As pointed out in Section 2, year-round concentrations of SO_4^{2-} (and NO_3^- in wintertime) may be representative for such large scales, but year-round concentration patterns of NH_4^+ and NO_3^- in summertime are determined more locally. In addition, assumptions on spatial homogeneity (e.g., land use and roughness characteristics) do not hold for determining large-scale deposition. In the case of particle deposition, the process is treated rather superficially using a constant value for v_d (EMEP) or r_c (TREND).

To account for spatial variability, the use of "filtering factors" has been proposed to explain enhanced deposition to forests (Hettelingh et al., 1991). The magnitude of filtering factors currently applied for mapping deposition onto EMEP grid cells is, however, in conflict with the mass consistency of the EMEP model. One may also question the usefulness and validity of such factors and the way to obtain them.

For estimating deposition over regions, refinement of spatial scales is necessary to account for land use, make up of ecosystems within grid cells and surface roughness. On the national scale several attempts have been carried out in different countries (e.g., Erisman, 1993; Lövblad, 1992; UK Review Group, 1990). The main obstacle is how to incorporate reasonable values for particle deposition velocities. Based on information discussed above, deposition calculations should be made for grids not exceeding the size of 50 × 50 km^2 at most, considering the spatial scales of SO_4^{2-}, NH_4^+ and NO_3^- concentration patterns and land use characteristics (see also Erisman and Baldocchi, 1994; Erisman et al., 1993). The choice of 50x50 km^2 is consistent with practical considerations by balancing the desire of fine scaling, computational resources and accuracy of deposition estimates.

5.2. Local scale

On a small scale, considerable variation in deposition can occur due to variations in concentration and in landscape characteristics. For SO_4^{2-} and in some cases NO_3^-, the effect of local sources and resulting concentration variations on deposition will be of minor concern. For NH_4^+ and alkaline particles, however, local variations in concentration may be important. Roughness transitions (especially from relatively smooth terrain to forested areas) exert a large influence on dry deposition as has been demonstrated by throughfall measurements (Hasselrot and Grennfelt, 1987; Draaijers et al., 1988) and model calculations (Wiman and Ågren, 1985). Compared to throughfall measured well away from the edge, throughfall at the edge was enhanced by a factor of 1.2 to 3.9 depending on the compound involved. Although detailed explanations are not yet available, this enhancement appears to be affected by advection of pollution into the canopy at the forest edge, the deposition velocity of the component itself, canopy structure properties, increases in turbulence intensities and possibly other factors.

Particle deposition can also affect dry deposition of SO_2 and NH_3 and it thus contributes to the local scale variation of total dry deposition. The underlying mechanism is related to the pH dependent behaviour of both SO_2 and NH_3 dry deposition to wetted surfaces. Alkaline particles depositing on a surface may raise pH, which promotes SO_2 deposition and decreases deposition of NH_3. Deposition of particulate NH_4^+ could influence the air/water equilibrium of NH_3, resulting in a possible decrease of the NH_3 flux to the surface.

6. Synthesis and recommendations

6.1. Synthesis

Information on dry deposition of SO_4^{2-}, NO_3^-, NH_4^+ and alkaline particles derived from both models and experiments has been discussed in this paper. Our current knowledge of dry deposition, based on modelling as well as experiments, can be characterized as poor and inadequate for policy development. Many aspects of particle dry deposition remain uncertain, highlighted by discrepancies between measured and modelled results. In particular, some model results predict very low deposition velocities for acidifying particles. These results are, however, contradicted by a number of experimental results but the experimental uncertainties are large.

Therefore, we cannot accurately assess the contribution of dry deposition of particulate SO_4^{2-}, NH_4^+ and NO_3^- to total deposition fluxes. For alkaline elements, such as Na^+, K^+, Mg^{2+} and Ca^{2+}, particle dry deposition contributes significantly to the total deposition of these elements. However, it is difficult to give the magnitude of these fluxes. In the framework of critical load mapping attempts have been made (Hettelingh et al., 1991), but the data used are uncertain.

Particle dry deposition will certainly vary over Europe, but accurate, quantitative data are lacking. Some assessments of the deposition pattern over Europe have been performed, including particle dry deposition, but they carry large uncertainties. For particle dry deposition these uncertainties are probably even larger than for the dry deposition of related gaseous compounds. If the relatively high deposition velocities are valid, which are found in some measurements and which are not inconsistent with all models, particle dry deposition can be a significant contributor to the total deposition, particularly in West and Central European regions. Uncertainty in the dry deposition velocity of particles can be illustrated with recent results on a 5 × 5 km grid scale in the Netherlands and for a particular forest in more detail (Erisman et al., 1994; Ruijgrok et al., 1994). The forest location concerned a Douglas fir stand of about 20 m height with a surface roughness length of over 1.5 m. Modelling and measuring results obtained here indicate a yearly mean dry deposition velocity of around 1.2 cm/s for SO_4^{2-}. The uncertainty in the yearly mean level of v_d is estimated to be at least 65 % when calculated with a modified version of Slinn's (1982) model for this location. However, other schemes which can be used to calculate v_d give substantially different and lower values for the yearly averaged v_d of SO_4^{2-}: these estimates are between 0.2 to 0.6 cm/s. The range in mean values calculated for one location under identical conditions illustrates present inaccuracies in assessing particle dry deposition. Estimates for the dry deposition of acidifying particles onto forests on a 5 × 5 km scale in the Netherlands were 2–3 times higher than earlier estimates, which substantially increased the contribution of dry deposition of acidifying particles to total dry acid deposition.

6.2. Recommendations

Mapping of deposition fluxes should be carried out on the smallest scale possible, preferably with a maximum resolution of 50 × 50 km. A standard for the overall inaccuracy of both wet and dry deposition estimates must be set. Validation of estimates for particle dry deposition against measurements is necessary.

Data on air concentrations of particulate SO_4^{2-}, NH_4^+ and NO_3^- can be provided by EMEP and national networks, but the current spatial resolution (notably for NH_4^+ and NO_3^-) is inadequate. A short-cut could be provided by deriving concentration patterns from high resolution models, which necessitates accurate emission inventories (of both natural and anthropogenic sources) on a similar scale as well as accurate gas-to-particle conversion rates.

Current process oriented models for particle dry deposition are too complicated and uncertain for mapping dry deposition. Moreover, many of the input data (such as vegetation characteristics or particle size distributions) required by such models are not available on a routine basis on the spatial resolution recommended for mapping deposition. The most common approach in those cases will be to make calculations with fixed values for most input parameters, regardless of actual differences in surface and other conditions. In these circumstances such a process oriented model will behave like a bulk resistance model. Current frameworks used for mapping of deposition are based on a multiple resistance approach (Erisman, 1993). In order to obtain a common methodology for both particle and gas deposition and because of computational reasons it seems best to incorporate the description of particle deposition in a similar way in these model frameworks for mapping deposition. As yet, however, no well established bulk resistance models exist for particle deposition. The task will be to derive a parametrization of the surface resistance for particles which captures the main driving forces of deposition processes. The form of such a relationship should preferably be as proposed by Wesely et al. (1985) and Hicks et al. (1987). The relationship should be applicable in a wider context than the limited measuring conditions associated with present parameterizations.

Thus far, dry deposition of particles has received far less attention in research than the deposition of related gaseous compounds. As a consequence, many gaps exist in our knowledge about particle deposition, both experimentally and in modelling. Discrepancies exist between both methods, which necessitate further clarification. Little is known about the dependence of v_d on vegetation type, roughness characteristics, roughness transitions and stability conditions of the atmosphere. More work is required to address these influences adequately.

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