

Particle deposition to forests

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Abstract

Particle deposition to forest was studied using experimental and modelling results. Results show that the deposition of particles to forests has been underestimated until now. Particle deposition makes out reasonable contribution to the total deposition of acidifying components and base cations to forests. It was estimated that at Speulder forest the contribution of dry particle deposition to the total deposition was 18% for SO₄, 38% for NO₃, 23% for NH₄, 56% for Na, 47% for K, 69% for Ca and 65% for Mg.. Deposition of compounds via fog at the Speulder forest was estimated to be small (<5%). Results of the canopy exchange experiments at Speulder forest and of the comparison of atmospheric deposition estimates and throughfall estimates at Speulder show that net-throughfall fluxes and atmospheric dry deposition estimates are reasonably in agreement for all components, except nitrate. There is still large uncertainty in canopy exchange processes of oxidised nitrogen and in deposition estimates of the different gases contributing to the total oxidised nitrogen flux.

1. INTRODUCTION

In 1991, the results of the second phase of the Dutch Priority Programme on Acidification were reported in [1]. Although much knowledge had been gained during the approximately six years of research, some serious gaps remained. Therefore, a third limited phase was initiated to fill in the most important gaps [2]. One of the main uncertainties was recognized to be the estimation of atmospheric input of acidifying aerosols and base cations to ecosystems. Together with wet deposition, particle dry deposition is responsible for the atmospheric load to

ecosystems of compounds such as sulphate, nitrate, chloride and ammonium, base cations such as calcium, magnesium, sodium and potassium. Deposition of particles containing SO_4 , NO_3 , Cl and NH_4 contribute to the potential acidification and eutrophication (nitrogen components) of ecosystems. Compared to gaseous deposition of acidifying compounds, particle deposition velocities and fluxes are usually found to be small. However, it is believed that the dry deposition velocity of small particles and therewith the fluxes is currently underestimated for very rough surfaces, like forests [3-5]. Current knowledge is therefore insufficient to give an adequate assessment of the dry deposition of particulate sulphur and nitrogen over the Netherlands and Europe. There is need for quantification to evaluate critical load exceedances and abatement strategies for atmospheric pollution.

Base cation deposition may be of importance for nutrient cycling in soils and ecosystems and may also neutralise acid input. Base cation input is therefore important in the determination of critical loads and/or critical load exceedances for ecosystems. Ecosystems receiving a high atmospheric input of base cations have higher critical loads than those receiving smaller inputs [6]. Base cations are usually found in the coarse fraction of ambient aerosols. Their deposition velocities are therefore large.

In several studies where throughfall fluxes are compared with atmospheric deposition estimates, large differences between the two estimates for deposition have been found [7-9]. Establishing a link between the two is useful because it provides a way to estimate soil loads from atmospheric deposition estimates on the one hand, and it allows the use of the relatively simple and cheap throughfall method to determine atmospheric deposition on the other. The link between atmospheric deposition and soil loads is important because critical loads refer to soil loads and because atmospheric deposition estimates provide a link with emissions. Thus if critical load exceedances are used to estimate emission reductions, the relation between atmospheric deposition and soil load should be known. It was suggested in [8] that beside canopy exchange processes, aerosol and/or fog and cloud water deposition might be important processes contributing to the observed differences between atmospheric deposition estimates and throughfall measurements.

The three main research questions that have been addressed in this project are:

- what is the contribution of "acidifying" aerosols to the total acid input of nature areas?
- what is the relation between atmospheric deposition estimates and throughfall measurements and what is the contribution of aerosol deposition to the difference between the two estimates?
- how important is the coarse particle flux (base cations) for the nutrient cycle in nature areas?

The research needed to get satisfying answers to the three research questions was defined by a project group, in which research institutes and universities in the Netherlands participated with experience in the field of aerosol research. The work in the aerosol project is a joint initiative of the *National Institute for Public Health and Environmental Protection (RIVM)*, *KEMA (Laboratory for Environmental Research)*, *TNO (Institute of Environmental Sciences)*, *ECN (Netherlands Energy Research Foundation)*, *RUU-FG (Utrecht University, Department of Physical Geography)* and *WAU-AQ (Wageningen Agricultural University, Department of Air Quality)*. It was anticipated in the project that by executing all available experiments in combination with a large modelling effort, a more accurate estimation of particle dry deposition velocities for rough surfaces would be obtained. A model was selected from existing models of aerosol deposition to forests [10]. The model had to be representative for the Dutch situation (pollution climate). From model simulations, insight was gained to the most

important processes involved in aerosol deposition. The main processes were tested by experimental research at the Speulder forest research site. The results of the experiments led to a verification of the model description and a basis for a parametrisation of V_d in terms of routinely available data. The parametrisation is used for the generalisation of aerosol deposition to other nature areas in the Netherlands. In this paper, the main results of the project will be summarised. Several publications form the basis of the results reported here, these are listed in the reference list.

2. EXPERIMENTAL

The Speulder forest ($52^\circ 15' \text{ N}$, $5^\circ 41' \text{ E}$) is located at the Veluwe, a large undulating area with forests and heathlands in the central part of the Netherlands. The measuring site covers an area of 2.5 ha, planted with Douglas fir of the provenance Arlington. By the end of 1993 the trees were 33 years old. The canopy is well closed with the maximum leaf area density at a height of 10-14 m. The one-sided LAI was between 149 and 10 for the measuring years 1987-1993 [11]. The stem density varies between 765 and 1216 trees per ha. In 1993 the trees were approximately 22 m high [12]. The stand is surrounded by a larger forested area of approximately 50 km²; the nearest edge is at a distance of 1.5 km. The stand itself is surrounded by *Larix*, *Betula*, *Quercus*, *Pinus* and *Pseudotsuga* trees, with mean tree heights varying between 12 and 25 m. A small clearing of 1 ha is situated to the North of the stand. Large source areas of SO₂ and NO_x are located about 200 km to the south-east (industrial Ruhr area) and about 100 km to the south-west (Rotterdam port) of the 'Veluwe'. The forested area is surrounded by large agricultural areas with intensive livestock farming acting as sources of NH₃ due to volatilisation from animal manure.

The location Speuld was equipped with two towers and measuring facilities (Figure 1). One tower is used for gas deposition measurements [13-15]. A sonic anemometer mounted on the top of the scaffolding at 36.5 m height is used to measure the horizontal and vertical wind velocity, wind direction and friction velocity and the sensible heat flux. A net radiation meter and a temperature/relative humidity sensor are mounted 1.5 m outside the scaffolding towards the south, at 35 and 33 m height, respectively. The two boxes house the gas monitors, two pulsed fluorescence SO₂ analysers and two Luminox NO₂ analysers, these are used to measure the vertical concentration gradient at 36, 32, 28 and 24 m height. Ammonia concentration measurements are made at three levels (34.5, 32 and 28 m height) using continuous flow denuders [16]. Ozone concentrations were measured at four heights (35, 32, 28 and 24 m) using chemoluminescence monitors. At 35 m the eddy correlation flux of ozone was measured using Gsten ozone sampler and a Kaijo Denki sonic anemometer [15].

The second tower was completely used for the Aerosol project [17]. The tower is 36 m high and has a rectangle cross-section with a length and with of 2 and 1.5 m, respectively. During the experiments, meteorological data and dry deposition data for SO₂, NH₃ and NO₂ were available from the concurrent project on trace gas deposition [14,16]. The experiments conducted within the framework of this project are listed in Table 1. In addition to the measurements used for estimating deposition, concentrations of SO₄, NO₃, NH₄, Cl, Na, K, Ca and Mg were measured in two size classes as 24 hr averages. (< 2.5 µm and > 2.5 µm) during a period of nine months [18]. Concentrations of HNO₂, HNO₃, HCl, SO₄, NO₃, and NH₄ were determined during several days with annular denuders [19] and once every week as hourly

values with wet rotating denuders [20]. Canopy exchange research using labelled S compounds (S^{35}) has been performed by ECN [20]. Fog composition, fog droplet diameter and liquid water content were measured by KEMA and ECN during several measuring campaigns [18,20,21]. In this way all ingredients determining throughfall and atmospheric deposition were measured.

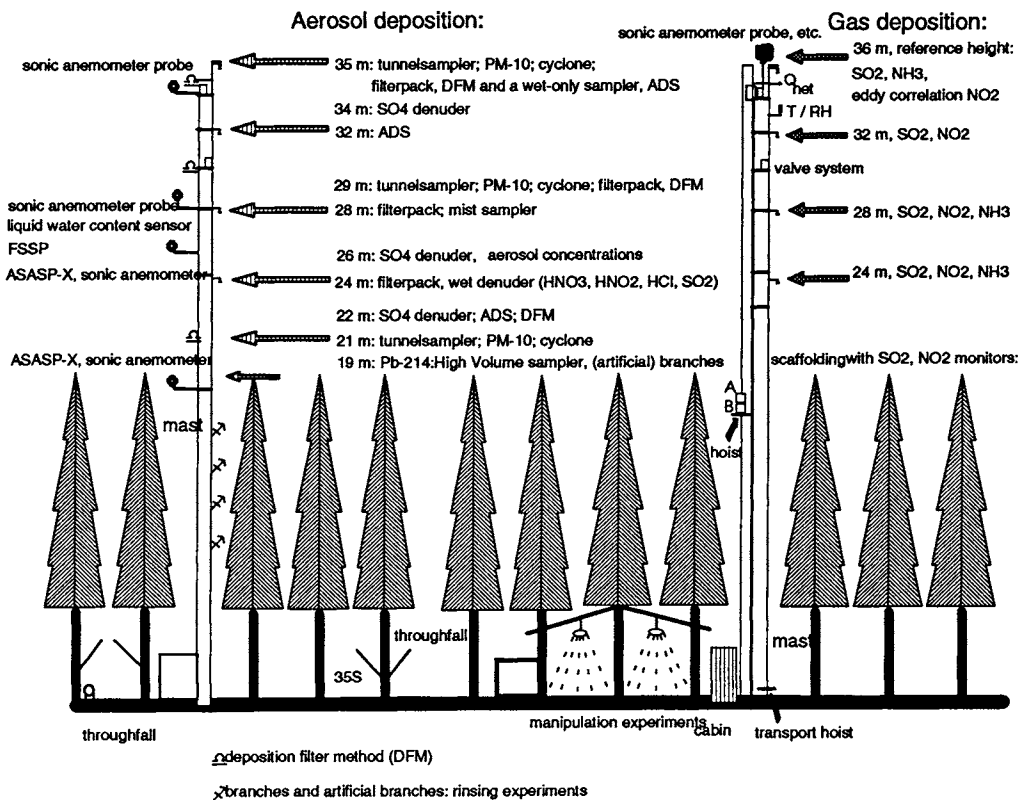


Figure 1. Schematic of the experimental set-up at Speulder forest.

The campaigns with the experiments were executed in December 7-12 1992 and during April 18 to May 10 1993. Throughfall, gas deposition and aerosol concentrations were measured continuously during a half year (26 November 1992 - 10 May 1993). The model used was that published by [25] and slightly modified by KEMA [10]. Parametrisations derived from model and measurement results were implemented in the RIVM models for estimating deposition of acidifying compounds at the Speulder forest and to nature areas in the Netherlands [5]. The experimental results were used to test and develop aspects of the model.

Table 1.

Overview of experiments performed at the Speulder forest to quantify particle dry deposition.

Technique	Species	Size range (μm)	Time resolution	Sampling heights (m)	Successful measurements	Error estimate flux (%)	Reference
Gradient	NO_3/SO_4	< ?	2 hrs	24, 30, 35	39	75	[22]
	$(\text{NH}_4)_2\text{SO}_4$	< ?	30 min	22, 26, 34	202	100	[20]
	Na, K, Ca, Mg	<2.5; <10; tot.	48 - 72 hrs	21, 29, 35	30	-	[19]
Eddy correlation	LWC	3 - 45	1 s	28	1637	20	[20]
	Fog droplets	1 - 95	15 min	28	1637	50	[20]
	Particles	0.1 - 3	0.1 s	18, 25	?	50 - 150	[23]
accumulation on branches	^{214}Pb	< 1	3 hrs	18	26	60	[20]
	$\text{SO}_4 \dots \text{Mg}$	0 - ∞	days	11, 15, 17, 19	13	60	[18]
DFM	Na, K, Ca, Mg	0 - ∞	7 days	21, 29, 35	14	>25	[19]
Throughfall	$\text{SO}_4 \dots \text{Mg}$	0 - ∞	max. 1 week	1.5	30	30	[24]
wet denuder	$\text{HNO}_3, \text{HNO}_2,$ HCl, SO_2	-	1 hr	24	1000	15	[20]
ADS	$\text{SO}_4 \dots \text{Mg}$	< 2.5	24 hrs	34	35	40	[19]
	$\text{HNO}_3, \text{HNO}_2,$ HCl, SO_2		12 hrs	21, 34	?	30	
aerosol sampler	$\text{SO}_4 \dots \text{Mg}$	<2.5, 2.5-15	24 hrs	26	~280	5	[18]

3. EXPERIMENTAL RESULTS COMPARED TO MODEL RESULTS

For each experiment model calculations were made using the modified Slinn model with forests characteristics estimated for the Speulder forest, size distributions from Table 2, actual meteorology and concentrations measured above the forest and the exact periods the experiments lasted. The individual model results are compared to measurements in [10]. The experimental work was aimed at evaluating different aspects or process descriptions of the model. Most processes involved in particle deposition differ in their dependency on particle size. The best way to evaluate individual processes is to use measurements representative for different particle size classes. However, as discussed in [10], the size distribution for which experiments are representative are uncertain. Furthermore, uncertainty in measurement results is usually so large that it is difficult to draw conclusions from a comparison. The experiments made here for evaluation of the modelled deposition mechanisms can be divided into four categories:

- ^{214}Pb measurements and eddy correlation measurements of particle deposition, representative for small particles with diameters smaller than $1 \mu\text{m}$
- NO_3 and SO_4 measurements which are representative for particles with a bimodal distribution, with most of the total mass in the size below $1 \mu\text{m}$
- measurements of total base cation deposition representative for large particles
- fog deposition measurements, representative for large particles.

Even though the MMD for which the four categories are representative differ much (Table 2), none of these is representative for a single deposition mechanism. All four are mainly determined by impaction and interception, and sedimentation in addition for fog deposition.

Table 2.

Component specific size distributions (mass median diameter, MMD, and geometrical standard deviation σ_g) derived from measurements in the Netherlands.

Component	MMD (μm)	σ_g
^{214}Pb	0.35	2.0
Ca	7.73	3.47
Mg	5.92	2.73
K	2.64	1.84
Na	5.12	2.64
SO ₄ < 2.5 μm	0.6	2.2
SO ₄ > 2.5 μm	4.5	1.6
NH ₄ < 2.5 μm	0.6	2.2
NH ₄ > 2.5 μm	4.0	1.6
NO ₃ < 2.5 μm	0.6	2.3
NO ₃ > 2.5 μm	4.5	1.6
fog droplets (December)	19.4	
fog droplets (February)	7.4	

The factor which is important for particle deposition, next to the size distribution of particles, is the friction velocity. If the size dependency and the u_* dependence of the dry particle deposition velocity is similar for the measurements and the model results, this might serve as some sort of validation of the most determining processes. Figures 2 and 3 show the u_* dependence of different experimental determined V_d and modelled V_d , respectively. In general the experimental and model results show a distinct influence of the size distribution and u_* on the dry deposition velocity of particles. Both figures show that $V_d(^{214}\text{Pb}) < V_d(\text{SO}_4) < V_d(\text{NO}_3) < V_d(\text{fog})$. Furthermore, the figures show similar relations between V_d and u_* for three of the categories distinguished above, although the variation in measured V_d per u_* class can be very high [10]. The category with base cations is not considered because no u_* dependent measurements were made.

The deposition velocity of fog is proportional to u_*^2 indicating that impaction is the most important process determining V_d . Furthermore, sedimentation is important. The V_d values of other compounds are proportional to u_* or a weak function of u_* ($\sim u_*^{1/2}$), indicating no distinct process is dominating rather a mixture of processes is occurring simultaneously [26]. Results are in line with other investigations, showing that particle deposition to forests can be considerable with V_d of several cm s^{-1} [27-29]. The relationships with u_* found for different particle size classes provide confidence in the experimental results. This can only be considered as indication, because errors in deposition estimates usually show a high correlation with u_* , because factors influencing the error in the deposition estimates highly correlated to u_* .

Evaluation of the integral model results can only be done with statistical parameters, including the uncertainty in modelled and measured values. The uncertainty in model results and the sensitivity of model results on input and model parameters has extensively been described in [10]. The overall error in modelled V_d integrated over the size distribution representative for acidifying aerosols is estimated about 65% [10]. For base cations this error is somewhat smaller (60%) because of the contribution of the relatively well parametrised description of sedimentation. The uncertainty in model estimates is lower than or about equal to the uncertainty in measurement results, with the exception of the fog deposition measurements which are estimated to have smaller errors (20%). The fractional bias of the

means (the relative difference between the mean calculated and observed value) falls within these limits. The relatively large sensitivity of the model and an inaccuracy of the same order in measuring results cause that a perfect 1:1 correspondence between both cannot be expected. Statistical testing of the difference between modelled and measured values is done using the non-parametric sign and Wilcoxon tests for paired samples. Both test revealed no significant differences between the mean values of modelled and measured fluxes or V_d 's [10]. This is of course mainly the result of the large standard deviations in measuring and modelling results.

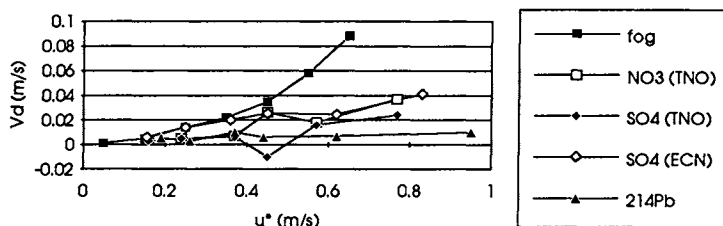


Figure 2. Experimental determined V_d versus u_* .

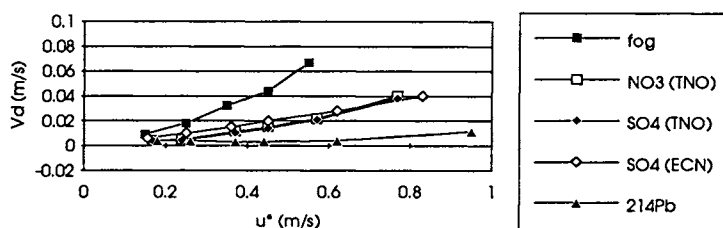


Figure 3. Summary of modelled V_d versus u_* .

It can be concluded that there are no strong indications for a significant underestimation or overestimation of the modelled V_d compared to measured values. The fact that the model is reasonably capable of describing a similar response of V_d to u_* as the measurements for different particle diameter ranges provides confidence in the process descriptions. Furthermore, it is concluded that particle V_d to forests and probably other rough surfaces are high. Average V_d values for half a year for fine particles at Speulder forest range from 1 to 2 cm s^{-1} (SO_4 , NO_3 , and NH_4) with daytime values being $1.3 \pm 1.2 \text{ cm s}^{-1}$ and night-time values being $1.0 \pm 1.4 \text{ cm s}^{-1}$ (SO_4). V_d values for coarse particles are about 5 cm s^{-1} with daytime values of $5.1 \pm 3.9 \text{ cm s}^{-1}$ and night-time values of $4.8 \pm 4.0 \text{ cm s}^{-1}$. In comparison, for the same period, V_d values for SO_2 , NH_3 and NO_2 were 1.5, 2.5 and 0.1 cm s^{-1} , respectively. This means that aerosol V_d to forest canopies in The Netherlands is underestimated until now with a factor of 2 to 3. For forests in Europe this is even higher, taking the EMEP model results [30]. It was estimated that at Speulder forest the contribution of dry particle deposition to the total deposition was 18% for SO_4 , 38% for NO_3 , 23% for NH_4 , 56% for Na, 47% for K, 69% for Ca and 65% for Mg. Deposition of compounds via fog at the Speulder forest was estimated to be small (<5%).

3.1 Parametrisation

The dry deposition velocity for particles was parametrised using the modified Slinn [25] model, for routinely available data. The general form for V_d at 50 m height is:

$$V_d = \frac{1}{\frac{1}{V_{ds}} + Ra(50)} \quad (1), \text{ where } V_{ds} \text{ can be estimated from: } V_{ds} = \frac{u_*^2}{u_h} E \quad (2)$$

u_h is the wind speed at canopy height, E is given for different components and conditions in [17]. Figure 4 shows parametrised and modelled values for the different z_0 values at Speulder forest. The comparison between modelled and parametrised values is very good, indicating that the most important parameters used for generalisation to other forests are well represented in the parametrisation.

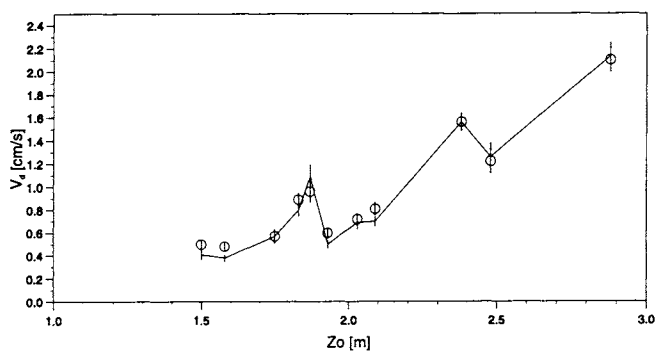
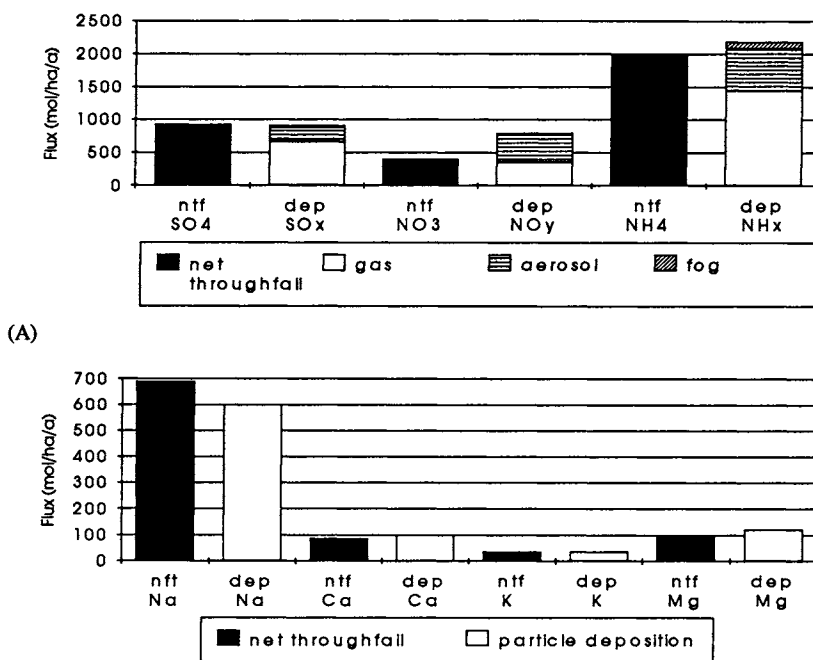


Figure 4. Parametrised (open circles) and modelled (line) V_d values (cm s^{-1}) as a function of z_0 for the whole dataset obtained at Speulder forest.

4. COMPARISON WITH THROUGHFALL

Net-throughfall fluxes were estimated using wet deposition and throughfall measurements during November 1992 to May 1993 [24,33]. Dry deposition for different components was estimated using the inferential technique with daily averaged measured concentrations and hourly averaged V_d calculated with the parametrisation given in section 3.2 for particles and those presented in [35] for gases. Comparison of net-throughfall estimates and dry deposition estimates are shown in Figure 5. Net-throughfall estimates were corrected for the influence of canopy exchange processes by applying the van der Maas/Ulrich model [31-33].



(B)
Figure 5. Net-throughfall estimates compared with dry and fog deposition estimates for acidifying components (A) and base cations (B) ($\text{mol ha}^{-1} \text{a}^{-1}$).

Figure 5 (A) and (B) show that net-throughfall estimates, corrected for canopy exchange, are in good agreement with dry and fog deposition estimates. The only exception to this is nitrate: these estimates are significantly different. It has been shown in [33] that most of the experiments at Speulder forest point in similar directions regarding canopy exchange. Although the estimates of the absolute amounts of components retained or leached in the canopy may differ depending on experiment, the average values give a good picture of the situation at Speulder forest. It shows that H^+ is taken up by the canopy, which is accompanied by leaching of K and to a smaller extent Ca and Mg. SO_2 taken up by stomata is eventually leached again, whereas NH_3 taken up via stomata is almost completely retained by the canopy. Oxidised nitrogen components are taken up by the canopy, especially NO_2 . Results of the experiments show that NO_3 uptake is negligible small, whereas NH_4 is taken up, in exchange with K, Mg or Ca. Na and Cl are considered inert. Highest uncertainty in canopy exchange estimates relates to estimates of the nitrogen components [33].

Observed differences between dry and fog deposition estimates from micrometeorological measurements and inferential modelling on the one hand and net throughfall fluxes on the other hand can not be regarded exclusively due to canopy exchange. Dry deposition estimates from micrometeorological measurements and inferential modelling are uncertain through errors in the air concentration measurements [34], their sometimes small time coverage and the uncertainties associated with the parametrisation of the dry deposition velocities [10,17]. Fog deposition

estimates are uncertain due to uncertainties associated with the estimation of water fluxes and the measurement of the average chemical composition of the fog droplets [21]. Uncertainties associated with the throughfall method when used for estimating dry and fog deposition include the dry deposition to the forest floor and understorey vegetation, dry deposition directly onto the throughfall gutters, the representativity of the throughfall sampling, the wet deposition estimate, the stemflow contribution and canopy exchange processes [8]. With canopy exchange processes being the only exception, mentioned factors probably contributed only to a very small extent to the uncertainty in the throughfall dry and fog deposition estimates in this study. Thus the combination of throughfall measurements and the empirical model lead to deposition estimates and soil load estimates which are at least as accurate as deposition estimates using other techniques. There is one drawback that is that the basic assumptions in the empirical model are not properly evaluated under different conditions (pollution climates, etc.).

6. CONCLUSIONS

The Slinn [25] model was selected from a number of models able to estimate particle deposition to forests. The model was used to determine the most important processes involved in the deposition of acidifying aerosols and base cations. The model formulation was slightly modified based on a comparison of different process descriptions used in the different models. Model results were compared with results of several experiments, comprising eddy correlation fog and particle measurements, vertical gradient measurements of sulphate, nitrate and base cations; and different accumulation experiments, such as ^{214}Pb measurements, leaf washing experiments, deposition plate measurements and throughfall measurements. Both model results and experimental results showed a strong dependence of the dry deposition velocity of particles on particle size and friction velocity. It was found that $V_d(^{214}\text{Pb}) < V_d(\text{SO}_4) < V_d(\text{NO}_3) < V_d(\text{base cations}) < V_d(\text{fog})$, in line with the size distributions. The deposition velocity of fog and base cations is proportional to u_*^2 indicating that impaction is the most important process determining V_d . Furthermore, sedimentation is important. The V_d values of other compounds are proportional to u_* or u_*^2 , indicating no distinct process being most important rather than a mixture of processes. It was concluded that the model results and measurement results are in good agreement. The model was used to derive a parametrisation of the deposition velocity of aerosols in terms of routinely available data. The parametrisation was used to generalize aerosol deposition to other nature areas in the Netherlands.

It was shown that deposition of fine particles is an important pathway for acid input to forests. It was confirmed that dry deposition velocities of particles to forests and probably other rough surfaces are high. Half year average V_d values for fine particles at Speulder forest ranged from 1 to 2 cm s^{-1} (SO_4 , NO_3 , and NH_4) with daytime values being $1.3 \pm 1.2 \text{ cm s}^{-1}$ and night-time values $1.0 \pm 1.4 \text{ cm s}^{-1}$ (SO_4). V_d values for coarse particles were about 5 cm s^{-1} with daytime values of $5.1 \pm 3.9 \text{ cm s}^{-1}$ and night-time values of $4.8 \pm 4.0 \text{ cm s}^{-1}$. In comparison, for the same period, V_d values for SO_2 , NH_3 and NO_2 were 1.5, 2.5 and 0.1 cm s^{-1} , respectively. These results led to the conclusion that the deposition of aerosols to forest canopies in The Netherlands is underestimated until now with a factor of 2 to 3.

Several field experiments were used to quantify canopy exchange. Using the new aerosol deposition estimates, differences observed between atmospheric deposition and throughfall fluxes of

acidifying compounds can almost completely be explained by canopy exchange processes. Uptake of H^+ and NH_4 is compensated for by leaching of Mg, Ca and most of all K. SO_2 taken up by stomata is eventually leached again, whereas NH_3 taken up via stomata is not leached from the canopy. Oxidised nitrogen components are taken up by the stomata in the canopy, especially NO_2 . Whether NO_3 is taken up is uncertain. Na and Cl are considered as inert. Highest uncertainty is found in the estimates of the nitrogen components.

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