G.J. Heij and J.W. Erisman (Editors). Acid Rain Research: Do we have enough answers?

Dry deposition of acidifying and alkaline particles to forests: model and experimental results compared

W. Ruijgrok

KEMA, P.O. Box 9035, 6800 ET Arnhem, The Netherlands

Abstract

This paper provides a comparison of modelling and experimental results on the dry deposition of acidifying and alkaline particles to forests. On average, a fair to good agreement is found between model and experimental results. Results show that the mean v_d of acidifying aerosol (such as SO_4) is around 1 cm s^{-1}, while for base cations (*e.g.* Ca) v_d is approx. 5 cm s^{-1}.

1. INTRODUCTION

In 1991, the second phase of the Dutch Priority Programme on Acidification ended [1]. A major uncertainty recognized was the atmospheric input of acidifying and alkaline aerosol to forests. The contribution of acidifying aerosol deposition to total input is unclear. Most commonly, the deposition velocity of such particles is believed to be small. However, over very rough surfaces it is poorly quantified and may be larger than assumed upto now [2-3]. In the 3rd phase of the Acidification Programme, a joint study was initiated to address the contribution of acidifying aerosol and base cations to atmospheric input and soil loads. This study consisted of a large experimental set-up together with a modelling study [4].

This paper compares estimates of particle deposition based on model and experimental results. The principal aim is to validate a model developed by Slinn [5] in such a way that deposition estimates derived from this model can be used for an assessment of the atmospheric input by particle deposition within the Netherlands. To this end a number of experiments were carried out in the course of 1992 and 1993 at the acidification research location Speulderbos in the Netherlands. The experiments differ in their set up, methods used, accuracy and each has its own advantages and disadvantages. Although results of all these experiments may not be directly comparable with each other, the whole array of results should bring about a clearer picture and understanding of particle deposition processes and of representative values for dry deposition velocities.

The model of Slinn was chosen following a review of current dry deposition models for forest conditions [6]. An extensive sensitivity analysis showed that the uncertainty in v_d may amount to 65% for acidifying compounds (60% for base cations). Some modifications were made: the influence of relative humidity on particle size and corrections for atmospheric stability were included. To compare modelled v_d with measured values, an integration of v_d was carried out over particle size distributions. Estimates for these distributions were made from concentration measurements carried Speuld or elsewhere in the Netherlands [6]. To model v_d a number of parameters characterizing the canopy are required; for the Speulder forest these were estimated from data measured or from literature values. To describe particle collection efficiencies, equations given in [7] were adopted instead of the original equations of Slinn, as they provided a better agreement with observational data. We will present here a selection of the comparison with measuring data only. A full description can be found in [6].

2. MODEL EVALUATION WITH MEASUREMENTS

Fog deposition

In December 1992 and February 1993 a number of fog events occurred during which eddy correlation measurements were taken of the turbulent deposition flux of fog droplets [8]. Model estimates of fog water deposition were made by integrating modelled v_d over the fog droplet spectrum which was measured. In December fog droplets showed a considerably larger mean diameter than in February. This will influence the contribution of sedimentation to the total flux of fog water droplets. Sedimentation was not measured directly but calculated from fog droplet spectra. Results show a large contribution of sedimentation to the total fog deposition (57% of the total flux in December versus 13% in February). Fig. 1 shows a comparison of modelled and measured time series. On the whole, there is a reasonable agreement between modelled and measured values with a notable exception on February 9. Model results show a clear relation of the turbulent deposition velocity with u_*^2 ($V_d = 0.20\ u_*^2$) which is close to the relation derived from measurements ($v_t = 0.195\ u_*^2$). A response of v_d to u_*^2 reflects the dominating influence of impaction.

^{214}Pb accumulation

Over a period of more than a year ^{214}Pb accumulation was measured [8]. The values obtained can provide an analogue for the deposition of acidifying particulate matter in the submicron size range (such as sulphate), because of a similar size distribution. We selected 26 measurements for comparison with model results; the other data may have been biased by interference of unattached ^{214}Pb. The experimental data relate to a local in-canopy deposition velocity at the measuring level, while the model provides a value referring to the entire canopy. To make the experimental data compatible with model estimates they have been scaled by a factor of 2.5. The uncertainty for this scaling factor ranges between about 1.5 and 4.

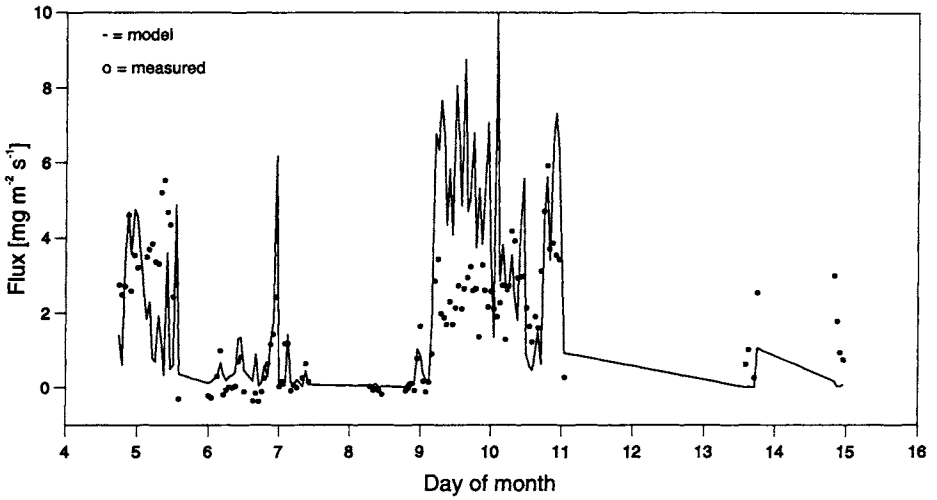

Figure 1 Time series of the deposition of liquid water over the Speulder forest in February 1993. The line indicates model results and circles measurements

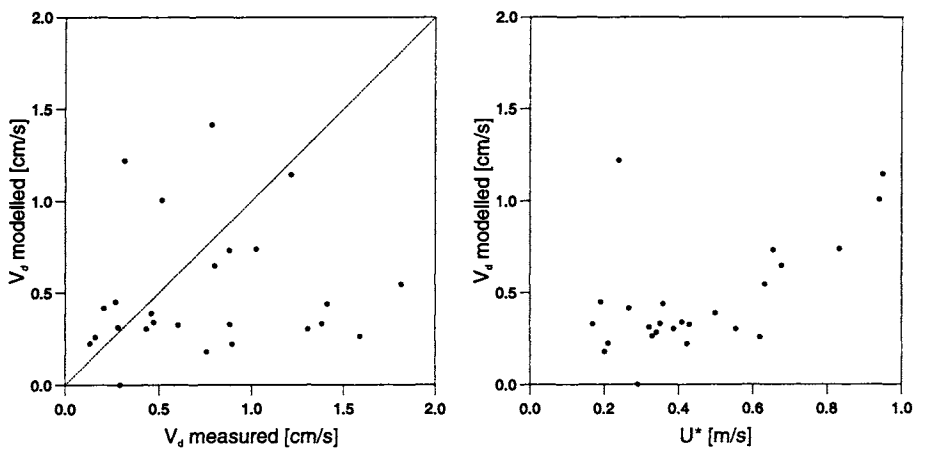

Figure 2 *(a)* Comparison of v_d measured (incorporating a scaling factor of 2.5) and v_d modelled. *(b)* The relation of v_d modelled with measured values of u_*

The model performs relatively poor in representing the deposition velocity of ^{214}Pb as measured in the canopy. This seems largely caused by two groups: one with a measured v_d between 1.3 and 2 cm s^{-1} for which the v_d is dramatically underpredicted; the other concerns a group with a calculated v_d of around 1.3 cm s^{-1} while measurements indicate a v_d of around 0.5 cm s^{-1}. This results in hardly any correlation between measurements and model results. We examined if two possible factors (relative humidity and friction velocity) may have caused

the highest deposition velocities measured, but no clear relations were found. Despite the small correlation, the average v_d calculated agrees quite well with the mean of the observations (although slightly underpredicted).

Concentration gradient measurements

A number of concentration gradient measurements (NO_3, SO_4 and particles) was carried out at the Speulder forest. All results were used for comparison with modelled values; however, we will present here results for SO_4 fluxes from thermodenuder gradient measurements. A relatively large dataset was available after selection, which shows a reasonable comparison with modelled fluxes below 0.3 μg m^{-3} (Fig. 3). Above this value, measured fluxes are systematically underestimated. Large positive and negative measured values seem to be the result of random measuring errors in individual concentration measurements. Model and measuring results show by and large a similar response of v_d to u_*, a driving force of deposition ($v_d \sim u_*^{1.2}$). Averaged model results of v_d fall within 95% confidence limits of the averaged measured data.

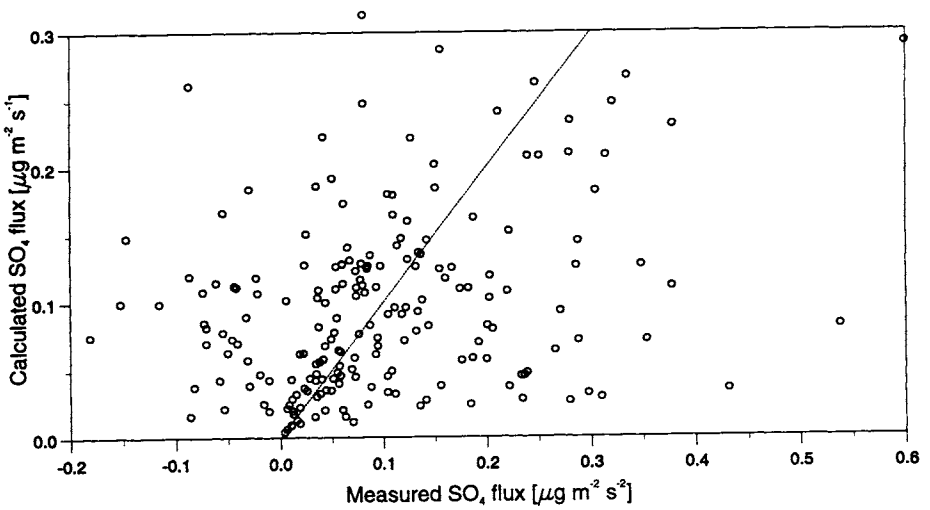

Figure 3 Comparison of modelled and measured SO_4 fluxes (thermodenuder results)

Throughfall

Draaijers *et al.* [9] made an analysis of throughfall fluxes and canopy exchange processes at Speuld. As part of this analysis atmospheric deposition estimates were made for the various acidifying and base cation compounds. A summary of results is presented in Table 1. To assess the performance of the particle deposition model, the components in throughfall most suitable for comparison are Na and Cl. The dry deposition of these two elements is via particle deposition only (as opposed to the sulfur and nitrogen compounds),

while canopy exchange processes do not influence their throughfall fluxes significantly.

To establish the atmospheric input by particle dry deposition we inferred values of v_d for acidifying and alkaline particles using the modified model of Slinn and continuous meteorological data measured at Speulder forest. Size distributions as derived from measurements at Speuld [6] were applied to obtain an integrated value of v_d. Fluxes were calculated from these integrated v_d values and the series of aerosol concentration measurements [10]. Results are shown in Table 1 for the period November 1992 to May 1993. The estimates are based on a time coverage of around 65%. Estimates of the input by dry gaseous deposition and by fog have been derived from gradient measurements and inferential modelling [4,8,11].

Table 1
Estimates of total atmospheric input (consisting of gaseous dry deposition, aerosol dry deposition and fog deposition) at Speulder forest. For comparison, net throughfall fluxes as well as net throughfall corrected for the canopy exchange processes are indicated. Fluxes are in mol ha^{-1} yr^{-1}. Also shown are mean and standard deviation of aerosol v_d used in these calculations (cm s^{-1})

	SO_x	NO_y	NH_x	Na	Cl	Ca	K	Mg
• gas	663	356	1443	0	0	0	0	0
• aerosol	216	414	645	599	855	101	34	118
• fog	34	23	96	2	4	1	1	0
total atmospheric input	913	793	2184	599	889	102	35	118
uncertainty	*40%*	*40%*	*50%*	*50%*	*50%*	*50%*	*50%*	*50%*
net throughfall	924	394	1728	692	802	159	305	138
net TF - canopy	924	394	1983	692	802	86	35	98
exchange	*30%*	*40%*	*40%*	*30%*	*30%*	*40%*	*50%*	*40%*
uncertainty								
average v_d	1.3	1.9	1.0	5.0	5.0	5.7	2.8	5.8
standard deviation	1.5	2.4	1.1	4.1	4.1	4.4	2.9	4.5

The long-term atmospheric deposition estimate of SO_x nearly equals the net throughfall flux measured. For Na and Cl, there is only a small difference between both ways of estimating input. In the case of NH_x and NO_y and most base cations, total atmospheric deposition is lower than net throughfall. However, if a correction is applied for the influence of canopy exchange processes atmospheric deposition and net throughfall fluxes compare reasonably well in the case of NH_x and base cations. Corrected net throughfall fluxes of Ca and Mg are slightly smaller than the atmospheric deposition estimate. However, this is probably caused by an overestimation of the canopy leaching of Ca and Mg by the canopy exchange model [9]. On the whole, long-

and SO_4 measurements for which the largest data sets were available. For these two compouds, also a reasonable simulation of the time series could be made. Moreover, a comparison of model and measuring errors gives no strong support for a significant biases of the deposition velocity by the model used.

Table 2
Model performance to simulate v_d measured with various techniques at Speulder forest. Four dimensionless indicators of model performance are presented: correlation coefficient (R), fractional biases of mean (FBM) and variances (FBV) and normalized mean square error (NMSE). For comparison, mean and standard deviation (in cm/s) are shown as well for model and measuring results

Intercomparison	R	FBM	FBV	NMSE	v_d model	σ model	v_d meas.	σ meas.	n
Fog	0.57	0.18	0.71	1.12	2.9	1.8	2.8	2.3	116
^{214}Pb	0.15	-0.37	-0.66	1.01	0.5	0.3	0.7	0.5	26
NO_3 (filterpack)	0.55	0.02	-1.06	1.75	1.2	1.1	1.2	1.9	23
SO_4 (filterpack)	0.42	0.55	-0.60	2.66	1.1	1.0	0.7	1.4	23
SO_4 (thermodenuder)	0.33	-0.08	-1.54	1.94	2.1	1.2	2.3	3.4	169
Ca (DFM, unscaled)	0.78	1.44	1.86	5.16	4.1	2.5	0.8	0.5	14
Ca (DFM, scaled)	0.78	0.62	0.93	0.59	4.1	2.5	2.4	0.9	14
Na (throughfall)	0.52	-0.14	-0.10	0.26	5.0	1.5	-	-	8
SO_4 (throughfall)	0.77	0.01	0.52	0.04	1.3	4.1	-	-	8

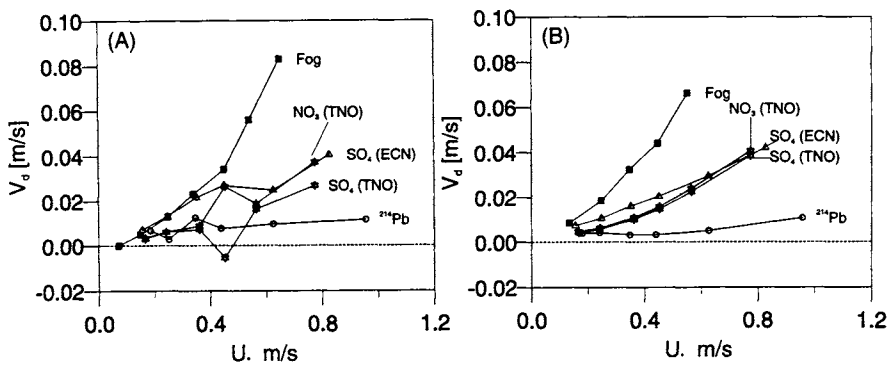

Figure 4 A summary of the averaged measured (A) and modelled (B) v_d values as a function of averaged values of u_* (in intervals of 0.1 m/s)

From our comparison one can obtain little ground to reject model results, although model performance is not very satisfying. However, present outcomes confirm earlier observations of higher deposition velocities of acidifying particles onto forests and other very rough surfaces. This does not hold only for the deposition measurements carried out in Speuld, but also for the model

term averaged particle deposition fluxes seem to be in line with input estimates obtained from (corrected) net throughfall. This provides confidence in the long-term averaged values of v_d calculated for particle dry deposition as well as in the performance of the canopy exchange model.

In order to make an evaluation on shorter time scales atmospheric deposition estimates were compared with net throughfall fluxes of 16 individual sampling periods (varying in duration from 57 to 493 hours). For these 16 individual events significant relationships between atmospheric deposition estimates and net throughfall fluxes could not be detected. This can largely be explained by incomplete wash-off of dry deposition from the canopy by precipitation [9]. This incomplete wash-off created a time dependency of throughfall samples on previously collected samples. A second reason for a lack of correspondence is the limited coverage in time of measurements on which some of the atmospheric deposition estimates are based. The limited time coverage can potentially introduce large errors in deposition estimates. Enlarging the averaging period over which deposition is calculated (by taking two consecutive periods together), modelled fluxes and net throughfall agree reasonably well (see results for Na and SO_4 in Table 2).

3. SYNTHESIS

We presented a comparison between deposition flux measurements of various compounds and tracers with estimates based on calculations with Slinn's dry deposition model. A summary of results can be found in Table 2. The model version used for these calculations included alternative descriptions of particle collection efficiencies [7], instead of Slinn's original equations. These alternatives were adopted as they yielded significantly higher deposition velocities. Such differences reflect the uncertainty in current understanding of the particle deposition process. The option of adopting alternative efficiency descriptions was chosen as it seemed at first hand to match more closely with the data measured.

To assess the performance of the model we used four different indicators to measure correspondence between measured and modelled fluxes. None of these indicators point at a good agreement; at best, their values illustrate a reasonable performance due to the large scatter. The cause for this are the relatively large inaccuracies of measuring methods used and the sensitivity of the model which is of the same order (60 - 130%). Depending on the compound, the averaged modelled fluxes differ with 2% to 144% from the averaged measured fluxes. The largest difference applies to a comparison with dry deposition fluxes from a filter method, a technique which may be characterized as unreliable. With respect to reproducing the variability in measured fluxes, the model seems to underestimate this variability systematically. On the other hand, the model adequately reproduces the response of v_d to u_* (an important driving force of particle deposition) by showing similar features as the measurements (see Fig. 4). This holds in particular for the case studies with fog

simulations. They also support the adjustment by Erisman [11] of the original parametrisation of v_d by Wesely *et al.* [12] in order to obtain higher values for acidifying particles. With respect to Slinn's deposition model, such higher values can only be calculated if alternative descriptions of particle collection efficiencies are used.

The present, adjusted version of Slinn's deposition model can in principal be useful to generalize dry deposition of acidifying particles to forests and other rough surfaces. A more convenient way (because of computational reasons) is to rely on a parametrisation of v_d with u_*. Secondly, as most input parameters of the model are not available for making deposition estimates for the entire Netherlands and also detailed particle size data are lacking, the model response will be primarily depending on u_*. Such a relation has been successfully derived from this model [4,6]. A parametrisation will, no doubt, cause inaccuracies and extrapolations have to done with caution, but, at the same time, these inaccuracies will be mostly due to the uncertainties of the model. Using the model itself will hardly reduce uncertainty in v_d for such purposes. It may only make the sources of such uncertainties more explicit.

4. REFERENCES

1 Heij G.J. and Schneider T. (1991). *Studies in Environmental Sciences 46*, Elsevier, Amsterdam.

2 Ruijgrok W., Davidson C.I., and Nicholson K.W. (1994). Dry deposition of particles: implications and recommendations for mapping of deposition over Europe. Accepted for publication in *Tellus*.

3 Nicholson K.W. (1988). *Atmos. Environ.* **22**, 2653-2666.

4 Erisman, J.W., Draaijers G., Duijzer J., Hofschreuder P., Van Leeuwen N., Römer F. Ruijgrok W. and Wyers P. (1994). *Particle deposition to forest.* Elsewhere in this volume.

5 Slinn W.G.N. (1982). Predictions for particle deposition to vegetative surfaces. *Atmos. Environ.* **16**, 1785-1794.

6 Ruijgrok W., Tieben H. and Eisinga P. (1994). KEMA, Arnhem, report 20159-KES 94-xxxx.

7 Wiman B.L.B., and Ågren G.I. (1985). *Atmos. Environ.* **19**, 335-362.

8 Wyers G.P., Veltkamp A.C., Vermeulen A.T., Geusebroek M., Wayers A., Möls J.J. (1994). ECN, Petten, report ECN-C-94-0xx.

9 Draaijers G.P.J., Erisman J.W., Van Leeuwen N.F.M., Römer F.G., Te Winkel B.H., Wyers G.P. (1994). RIVM, Bilthoven, report 712108004.

10 Römer F.G. and Te Winkel B.H. (1994). KEMA, Arnhem, report 63591-KES/MLU 93-3243.

11 Erisman J.W. (1993). *Water, Air and Soil Poll.*, **71**, 51-80.

12 Wesely M.L., Cook D.R., Hart R.L., and Speer R.E. (1985). *J. Geophys. Res.* **90**, 2131-2143.