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

Deposition of aerosol to coniferous forest

G.P. Wyers, A.C. Veltkamp, M. Geusebroek, A. Wayers, J.J. Möls

Netherlands Energy Research Foundation ECN,
P.O. Box 1, 1755 ZG Petten, The Netherlands

Abstract

The deposition velocity of ammonium(bi)sulphate (MMD 0.8 µm, σ_g 2) was derived from vertical concentration gradients using flux-profile relationships. The deposition velocity has a near-linear dependency on u_* and an average value of 0.012-0.015 m s^{-1}. The dry deposition of ^{214}Pb was investigated by measuring airborne activity and activity accumulated on Douglas fir branches. This radioisotope is a natural tracer for submicron aerosol (AMD 0.4 µm, σ_g 3). The correlation of V_d with u_* is poor. The average deposition velocity ranges from 0.0061-0.0110 m s^{-1}, with a best estimate of 0.0073 m s^{-1}, a factor 1.6-2.1 smaller than the deposition velocity for sulphate. The difference is regarded to be due to the difference in size distributions of ^{214}Pb and sulphates.

1. INTRODUCTION

Wind tunnel experiments have shown that the dry deposition of aerosol is strongly dependent on particle size [1]. Because these studies indicated very small deposition velocities less than 1 mm s^{-1} for particles in the range 0.1-1 µm, the deposition of submicron aerosol has been considered relatively unimportant, as compared to deposition of gaseous compounds. It has been reported however by Hicks et al. [2] that deposition velocities for submicron aerosol can be considerably larger than 1 mm s^{-1} over vegetative surfaces in the field. Few measurements of aerosol deposition have been made over forests. Hicks et al. [3] measured sulphate fluxes over a deciduous forest by eddy correlation. They found that V_d ranged from virtually 0 at night to a maximum of 1 cm s^{-1} at daytime with a long-term average value of 0.6 cm s^{-1}. Similar values are reported by Wesely et al. [4] for a pine forest: the deposition velocity ranges from 0.90 cm s^{-1} for moderately unstable conditions to 0.48 for a neutral atmosphere.

In order to estimate the deposition velocity of submicron aerosol over forests, two experiments were performed at the site Speuld, located in the center of The Netherlands. This site is a 2.5 ha monoculture of Douglas fir with a mean tree height of 20m. The canopy is dense and had in 1992 a maximum one-sided Leaf Area Index (LAI) of 10.7 (Jans and Steingröver, pers.comm.). The deposition velocity of ammonium(bi)sulphate was determined from vertical concentration gradients measured above the canopy. The natural radio-isotope ^{214}Pb which occurs attached to aerosol surfaces was used as a tracer for submicron aerosol. The deposition velocity of this radon daughter was determined by counting the airborne activity and the activity deposited on the needle surfaces.

2. DRY DEPOSITION OF SULPHATES

2.1. Experimental

Vertical concentration gradients of ammonium(bi)sulphate and sulphuric acid over the forest were measured by CuO-coated thermodenuders with a time resolution of two hours. Three instruments were placed on a scaffold at heights of 22m, 26m and 34m above the forest floor. Before starting the measurements over the forest, three thermodenuders were run simultaneously in Petten for a period of two weeks to examine systematic and random errors in the determination. The results indicate systematic differences up to 6%. All measurements were corrected for the detected bias on the basis of this intercomparison. After correction, the average of the relative standard deviation of the triplicate measurements amounted to 4%. Turbulence intensity and sensible heat flux were measured by eddy correlation (Erisman, pers. comm.).

2.2. Results

From 17 May to 23 August 1993 measurements of the vertical concentration gradient were performed every two hours. The average concentration of ammonium(bi)sulphate in the period May - August 1993 was 4.8 ± 3.2 $\mu gSO_4\ m^{-3}$. The sulphuric acid concentration was very low, mostly below 0.2 $\mu gSO_4\ m^{-3}$ and never exceeding 0.8 $\mu gSO_4\ m^{-3}$, indicating a very high degree of neutralization of sulphate by ammonia.

Only the measurements collected in the period 23 July - 23 August were used to calculate deposition velocities for sulphate aerosol, since these were collected while all equipment was operating properly. For the calculation of c_* modified flux-profile relations were applied, which have been derived for this site from a comparison between heat fluxes directly measured by eddy correlation and calculated from temperature gradients by Bosveld [5]. A more recent study by Weststrate and Duyzer [6] involving a comparison of eddy correlation measurements of fluxes of ozone and sensible heat with vertical profiles of ozone and temperature yielded similar results. C_* was calculated by linear regression of c(z) versus the logarithm of the stability-corrected height, with the correction factor α calculated from the effective height z_{eff} [5], using a displacement of 15m and a reference height z_r of 23.5m [6]. The deposition velocity V_d and the surface deposition velocity V_{ds} are calculated for the lowest measurement height $z = 22m$. The surface deposition velocity V_{ds} is obtained from V_d by subtraction of the contribution of turbulent transport and is defined as $(V_d^{-1}-r_a)^{-1}$ [4].

2.3. Discussion

A time series of V_d, V_{ds} and u_* is given in Figure 1 for the period 23 July - 2 August 1993. Despite the large scatter in V_d, a weak positive correlation can be observed with the friction velocity. The scatter in V_{ds} is considerably larger than in V_d.

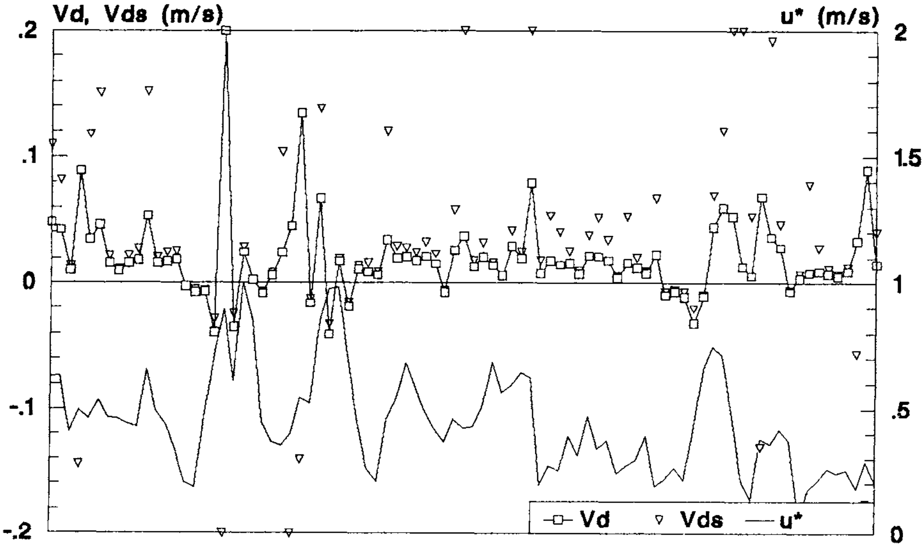

Figure 1. *Time series of deposition velocity (V_d), surface deposition velocity (V_{ds}) and friction velocity (u_*) above the Speulder Forest, 23 July - 2 August 1993.*

A random error of 4% in the individual concentration measurements may, under neutral atmospheric conditions, correspond with an uncertainty in V_d of 1.5 cm s^{-1}. Assuming that the nature of the errors is stochastic, the uncertainty can be reduced by averaging the deposition velocities for ranges of u_*, provided u_* is the determining factor. As the uncertainty in the individual gradient measurements is high, all measurements were sorted on u_*, and for each u_*-range a 50-percentile and an average V_d were calculated. Initial selection of the data on sufficient homogeneous fetch and u > 1 m s^{-1} yielded 339 measurements for further processing (Table 1).

Both $V_d(50)$ and average V_d display a positive correlation with u_*-class, but the average is a factor two larger than the 50-percentile of V_d, which is caused by a few extreme values. This is obvious from the large standard deviation which is mostly exceeding the value of the average deposition velocity. The true V_d is therefore better approximated by the median than by the average. The $V_d(50)$ increases with increasing friction velocity and can be approximated by the power function:

$$V_d(50) = 0.0526\, u_*^{1.64} \qquad \text{with } r^2 = 0.988 \qquad (1)$$

At low values of u_* (<0.4 m s^{-1}) however approximately 25% of the measurements yield apparent upward fluxes, which probably cause an underestimation of V_d at low friction velocities. Sorting of the measurements was also attempted using rh or z_o as a key. Sorting on rh yielded a poor, inverse relationship with $V_d(50)$, which is due to the fact that high relative humidities occur at night, when turbulence intensity is low. A similar problem was encountered with sorting on z_o, since the roughness length is dependent on wind direction, which is at this site strongly correlated with wind speed. It can be concluded that u_* is the dominant parameter determining the magnitude of the deposition velocity.

Table 1. *Deposition velocities for different u_*-ranges. Selection on u (> 1 m s^{-1}) and sufficient fetch*

u_*-range m s^{-1}	$\bar{u}_*$ m s^{-1}	$V_d(50)$ m s^{-1}	V_d m s^{-1}	s m s^{-1}	n
0.0-0.1	0.058	0.0005	0.0009	0.0012	28
0.1-0.2	0.153	0.0021	0.0049	0.0016	64
0.2-0.3	0.245	0.0059	0.0139	0.0437	71
0.3-0.4	0.348	0.0092	0.0300	0.0600	50
0.4-0.5	0.449	0.0184	0.0261	0.0349	53
0.5-0.7	0.610	0.0196	0.0306	0.0462	42
0.7-1.0	0.822	0.0365	0.0584	0.0827	31
all data	0.357	0.0062	0.0216	0.0480	339

Changes in concentration with time may result in artefact upward or downward fluxes. A further selection was made by setting the maximum error due to storage changes at 10% of the flux ($\delta c/\delta t\ (z_2-z_1)\ F^{-1} < 0.1$) [7]. This selection also served to eliminate spurious concentration measurements. After this selection 217 data remained for further evaluation. A further selection was aimed at removal of unrealistically high deposition velocities by limiting the maximum value of $|V_d|$ to 2 r_a^{-1}, i.e. two times the maximum deposition velocity for momentum. This led to disqualification of only 15 more measurements. The results are listed in Table 2.

The median values for $u_* \leq 0.4$ m s^{-1} have become higher compared to the first selection which was based on windspeed and fetch only (Table 1). This increase in V_d is partly the result of elimination of negative values resulting from poorly measured concentration gradients. The number of measurements resulting in apparent upward fluxes is now reduced to 15%. A possible bias towards higher deposition velocities could have been caused by removing measurements with storage errors exceeding 10% of the flux, since measurements representing small fluxes are preferentially removed. Both the elemination of apparent upward fluxes and the selection on storage errors lead therefore to higher values of $V_d(50)$, especially at low to moderate friction velocities (≤ 0.4 m s^{-1}, see Tables 1 and 2). Therefore, the true value of the deposition velocity for $u_* \leq 0.4$ m s^{-1} lies probably in between the values listed in Tables 1 and 2.

Table 2. *Deposition velocities for different u_*-ranges. Same selection criteria as in Table 1, with $\delta c/\delta t\,(z_2-z_1)\,F^{-1} < 0.1$ and $|V_d| < 2\,r_a^{-1}$*

u_*-range $m\,s^{-1}$	$\bar{u}_*$ $m\,s^{-1}$	$V_d(50)$ $m\,s^{-1}$	$\bar{V}_d$ $m\,s^{-1}$	s $m\,s^{-1}$	n	$V_{ds}(50)$ $m\,s^{-1}$	$\bar{V}_{ds}$ $m\,s^{-1}$	s $m\,s^{-1}$
0.0-0.2	0.156	0.0044	0.0071	0.0140	20	0.0034	0.0057	0.0403
0.2-0.3	0.248	0.0088	0.0132	0.0219	40	0.0108	0.0114	0.0988
0.3-0.4	0.345	0.0178	0.0216	0.0276	32	0.0092	-0.0062	0.2259
0.4-0.5	0.450	0.0211	0.0270	0.0313	46	0.0217	0.0340	0.2799
0.5-0.7	0.615	0.0196	0.0248	0.0344	38	0.0239	0.0370	0.1353
0.7-1.0	0.834	0.0344	0.0399	0.0557	26	0.0292	0.0697	0.1261
all data	0.400	0.0158	0.0227	0.0341	202	0.0167	0.0256	0.1865

Average and 50-percentile of the deposition velocities are now in reasonable agreement. The standard deviations have decreased and are of approximately the same magnitude as the average deposition velocities. The 50-percentile of the surface deposition velocity V_{ds} is similar to that of the total deposition velocity V_d, but the average V_{ds} is highly variable and has a standard deviation several times its value.

$V_d(50)$ and $V_{ds}(50)$ both show a high positive correlation with u_* (Fig. 2). Linear regression as a function of average u_* for each range yields:

$$V_{ds} = 0.0380\,u_* - 0.0004 \qquad \text{with } r^2 = 0.895 \tag{2}$$

$$V_d = 0.0400\,u_* \qquad \text{with } r^2 = 0.903 \tag{3}$$

This near-linear relationship between deposition velocity and friction velocity is somewhat unexpected. When impaction is the only mechanism responsible for passing the viscous sub-layer, V_d should increase stronger than linearly with u_* [8]. The observed linear relationship suggests that (a combination of) other mechanisms is responsible for this dependency on u_*. Model calculations yielded an analogous near-linear relationship between Vd and u_* [9], which support the validity of (3).

For V_d = 0-0.05 m s^{-1} much better agreement between observed and predicted deposition velocities was obtained using relationship (3) rather than (2). The results given in Figure 3 show a reasonable correlation for V_d between 0 and 0.05 m s^{-1}, but also indicate large positive and negative values up to ± 0.1 m s^{-1} which deviate strongly from the predicted behaviour and are very likely the result of erroneous concentration gradient measurements. Despite the possibility that deposition velocities at $u_* \leq 0.4$ m s^{-1} are slightly overpredicted (see above), equation (3) is considered the most accurate that can be derived from these measurements. Also shown in Figure 3 are deposition velocities predicted using parametrizations by Erisman [10] and Wesely et al. [4]. The latter was derived for sulphate

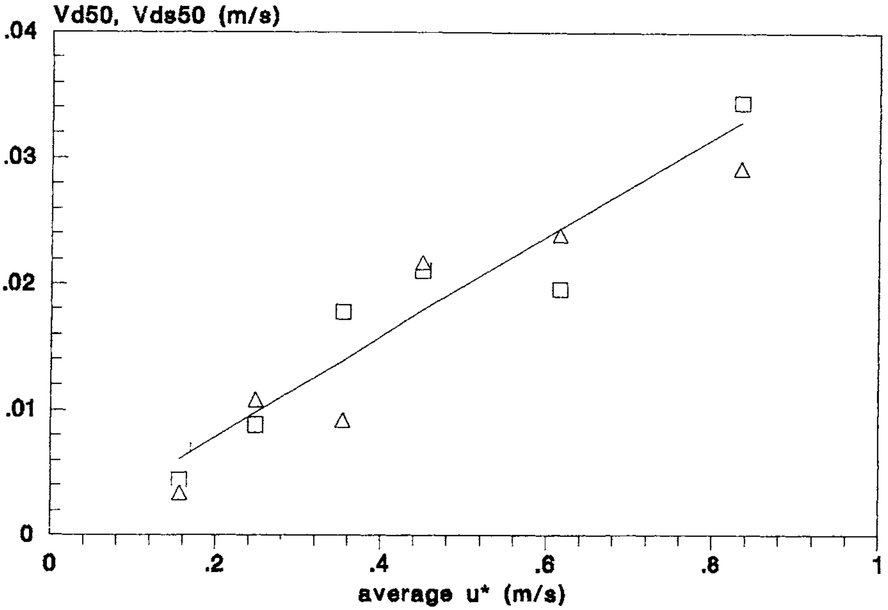

Figure 2. *Median deposition velocity (squares) and median surface deposition velocity (triangles) for SO_4 at different u_*-ranges as a function of average u_*. Selection criteria as listed in Table 2.*

deposition on grassland in the U.S.A.; the parametrization by Erisman is currently used to estimate aerosol deposition in the Netherlands. These parametrizations lead to an underprediction of the deposition velocities by approximately a factor two (Erisman) to ten (Wesely et al.).

Equation (3) was used to calculate average flux and deposition velocity from measured u_* and SO_4 concentration for the entire measurement period May-August 1993. This yielded an average deposition velocity of 0.0148 ± 0.0082 m s^{-1} and an average flux of -0.0738 ± 0.0618 μgSO_4 m^{-2} s^{-1}, corresponding to approximately 240 mol SO_4 ha^{-1} a^{-1}. For reasons given earlier this can be considered an upper estimate. A lower estimate can be obtained by applying equation (1), which yields an average deposition velocity of 0.0120 ± 0.0103 m s^{-1}, only 20% lower than that derived from (3), corresponding with an annual flux of approximately 200 mol SO_4 ha^{-1} a^{-1}.

The deposition velocities observed for sulphate aerosol over this forest are much higher than deposition velocities for submicron particles predicted from wind tunnel studies and are also high when compared to results from previous field studies over forests. For a deciduous forest with a similar tree height and a LAI at 50% of its summer maximum, Hicks et al. [3] found a long-term average deposition velocity of 0.6 cm s^{-1} with V_d/u_*

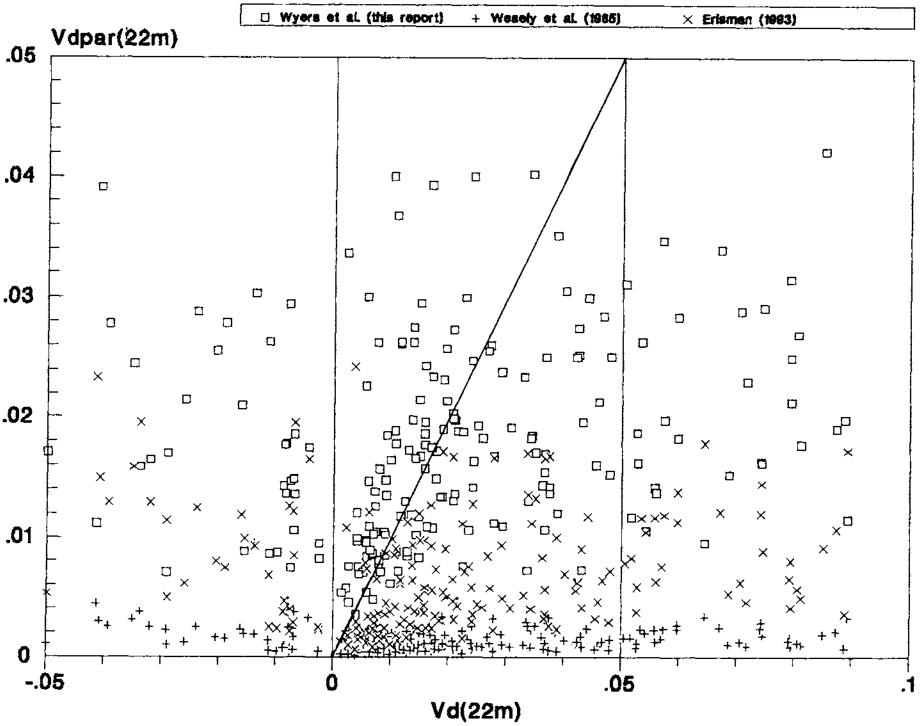

Figure 3. *Comparison of observed and predicted deposition velocities. Parametrizations from this report (equation (3)), Erisman [10] and Wesely et al. [4].*

ranging from 0.001 at stable conditions to 0.02 for a moderately unstable atmosphere. The average wind speed however was only 2 m s^{-1}. For the site Speuld an average of 0.04 is found for V_d/u_* at an average wind speed of 3.6 m s^{-1} (for the period May - August 1993). Dry deposition of aerosol in The Netherlands is currently estimated using a parametrization derived by Erisman [10], which calculates the deposition velocity as a function of friction velocity and atmospheric stability. Application of this parametrization to the measurement period May - August 1993 results in an average deposition velocity of 0.0062 ± 0.0051 m s^{-1}. The results from the present study indicate that dry deposition of sulphate on coniferous forest is underestimated by more than a factor two.

The deposition velocity is strongly dependent on particle size distribution, and it is possible that a few large particles lead to such high values. However, size distributions for antropogenic sulphate show that little mass is present as supermicron particles. Particle size distributions have not been measured during this experiment. In 1983 and 1984 size distributions of sulphate were determined with Andersen impactors during air pollution episodes in 1983 and 1984. The average can be represented by a log-normal distribution with a MMD of 0.8 μm and a σ_g of 2. Moreover, large particles > 5-10 μm will not pass the air inlet of the thermodenuders. A second point worth mentioning is the very dense canopy of this forest. To avoid interference with the measurements, thinning of this forest was postponed, and the one-sided LAI now amounts to approximately 11 (Jans and Steingrover, pers. comm.). The canopy may therefore have become a very efficient receptor surface for fine aerosol. A third possible reason for the high observed deposition velocities is hygroscopic growth of the particles in the humidity gradient above the surface. Growth is under most conditions very fast compared to vertical transport times [11] and may shift the size distribution near the receptor surface to particle diameters which deposit very efficiently.

3. DRY DEPOSITION OF ^{214}Pb

3.1. Experimental

^{214}Pb is a daughter isotope of ^{222}Rn, which itself belongs to the decay series of ^{238}U. After formation by decay of ^{226}Ra in the subsurface, ^{222}Rn may enter the atmospere, where it will decay according to the scheme:

$$^{222}\text{Rn (3.8d)} \rightarrow {}^{218}\text{Po (3.05min)} \rightarrow {}^{214}\text{Pb (26.8min)} \rightarrow {}^{214}\text{Bi}$$

After decay of radon, the daughter isotopes will react rapidly with trace gases and vapours forming clusters or will attach to existing aerosol surfaces within 1-100 s [12]. The attachment rate is a function of particle size. The daughter isotopes of radon can thus be used as a selective tracer to examine the deposition of aerosol onto surfaces such as vegetation canopies.

Fresh branches of Douglas Fir, collected near the measurement site, were mounted horizontally in a tower at 19 m height near the top of the canopy. After exposure for at least 3 h, twigs from the branches are rapidly (within 10 min) collected for radioactivity counting by gamma ray spectrometry. Airborne radioactivity is collected by sampling at 60 m^3/h on a 20x25 cm glass fiber filter using a high-volume sampler. The high-volume sampler is installed at the tower near the exposed branches mentioned above. The inlet of the sampler is positioned at 19.5 m height. After approximately two hours of sampling, steady state of ^{214}Pb on the filter is reached, i.e. the ^{214}Pb content on the filter is constant since collection and decay of this isotope on the filter are in equilibrium, assuming constant radioactivity levels in air during the sampling period. After steady state is reached, the filter is rapidly (within 10 min) prepared for radioactivity counting by gamma ray spectrometry.

From simultaneous measurements of the airborne activity collected on filters by high-volume sampling and the activity accumulated on needles at the top of the canopy, a "local" deposition velocity V_d(loc), representative for deposition on 1 m^2 of needle surface at 19 m near the top of the canopy, is derived for submicron aerosol following Bondietti et al. [13]:

$$V_d(\text{loc}) = \{L\ V_{air}\ E_{air}\ A_{needles}\}/\{S\ W\ E_{needles}\ A_{air}\}\ [\text{m/(s m}^2 \text{ needle area @ 19m)}]$$

L : decay constant ^{214}Pb
V_{air} : sampled air volume
E_{air} : counting efficiency gamma-measurement filter on 352 keV
$E_{needles}$: counting efficiency gamma-measurement vegetation on 352 keV
A_{air} : counts on filter, recalculated to t=0
$A_{needles}$: counts on vegetation, recalculated to t=0
S : specific leaf area (i.c. 6.0 m^2/kg dry weight)
W : dry weight vegetation sample

3.2. Results

The emission of radon from the earth's surface is a function of the radium content of the subsurface. Emanations of radon from the subsurface in the Netherlands are very low. Sufficient deposited activity for a reliable measurement was only obtained in air masses which had travelled a long distance over land. Measurements were thus mainly performed during easterly and southerly winds. In the period December 1992 - May 1994 42 measurements of airborne and deposited activity were performed at the site Speuld. Of these, 26 measurements corresponded with wind directions from the sector 90-180. These measurements were used for further evaluation. In Figure 4 V_d(loc) is shown as a function of friction velocity, measured above the canopy by eddy correlation.

Three measurements of the size distribution of ^{214}Pb, made at the site Speuld during easterly winds with a Sierra high-volume cascade impactor, confirmed that this isotope is associated with submicron particles. Assuming a log-normal size distribution, the Activity Median Diameter (AMD) and the geometric standard deviation σ_g were estimated at 0.4 µm and 3 respectively. According to Porstendörfer [12] the activity distribution of ^{214}Pb is unimodal and can be well approximated by a log-normal size distribution. Measurements of the activity distribution compiled by Porstendörfer indicate an AMD of 0.15-0.39 µm and a σ_g of 2.3-2.8. A comparison with size distribution measurements of antropogenic sulphate (MMD of 0.8 µm) indicates that the size distribution for ^{214}Pb is characterized by smaller particle diameters.

3.3. Discussion

In clean air masses from the North Sea or the Scandinavian countries anomalously high values were found for V_d(loc) (0.0068±0.0055 m/(s m^2 needle area). This was attributed to the presence of a large fraction of ^{218}Po and possibly ^{214}Pb in cluster mode (i.e., unattached to submicron particles) which deposits at a higher rate than accumulation mode aerosol [12, 14]. The attachment rate of radon daughters is related to the amount of aerosol surface present. In clean air masses the attachment rate will be considerably slower, resulting in a higher fraction of activity in cluster mode. Therefore only measurements in polluted air masses with (predicted) 36h-back-trajectories originating in the sector 90-180 were considered for studying the deposition velocity of fine aerosol.

The deposition velocities thus measured have to be corrected for the contribution by unattached activity. This correction is based on the following assumptions: an accommodation factor of unity, losses from recoil of 50%, a size distribution for sulphate aerosol as measured during air pollution episodes in 1982-1984 but with a reduction in

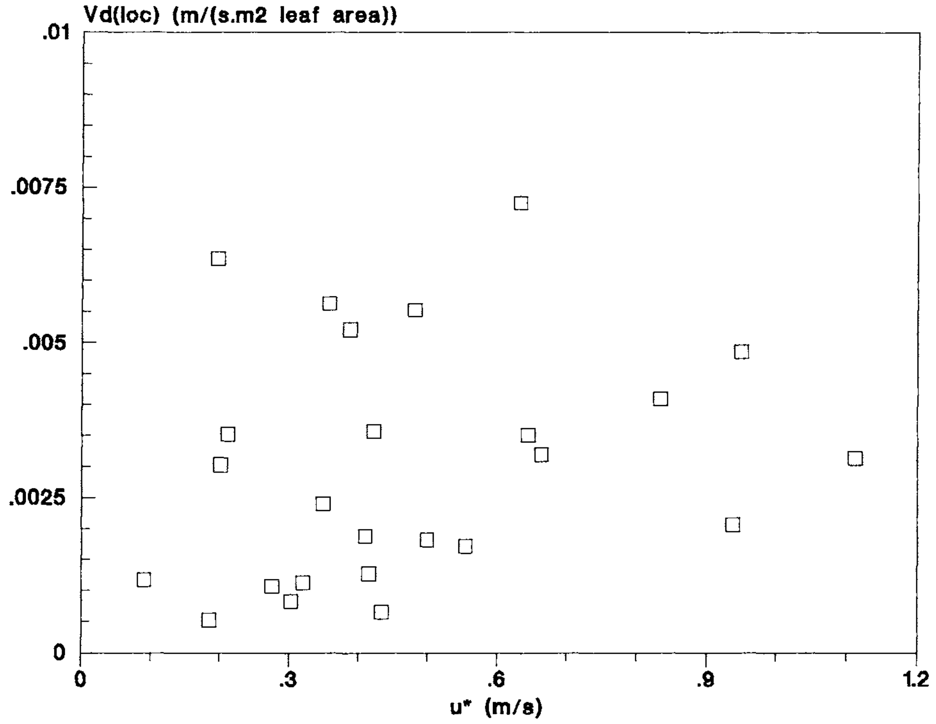

Figure 4. *The local deposition velocity of ^{214}Pb (representative for 1 m^2 needle area @ 19 m) as a function of u_* above the canopy.*

number concentrations of a factor two, and a diffusion coefficient for clusters in air of 0.05 $cm^2\ s^{-1}$. This yields an attachment rate of 8 $10^{-3}\ s^{-1}$. Furthermore taking a minimum mixing layer height of 500 m and a maximum deposition velocity for cluster mode aerosol of 0.05 $m\ s^{-1}$ (the average reciprocal aerodynamic resistance for this site) leads to a fraction of unattached activity of 1.25% or less. The deposition velocities for attached activity have been determined by subtracting the contribution due to 1% of unattached activity from the total deposition velocity. The V_d for cluster mode aerosol was calculated from $(r_a+r_b)^{-1}$. A diffusion coefficient of 0.05 $cm^2\ s^{-1}$ was used to calculate r_b. Brownian diffusion was assumed as the rate-limiting mechanism for transport of clusters through the viscous sub-layer, so possible electrical interactions were ignored. By this correction the deposition velocity for attached activity is decreased by 6% on average relative to the total deposition velocity.

The average V_d(loc) for attached activity of 26 measurements obtained from December 1992 to February 1994 was 0.00290 ± 0.00194 $m\ s^{-1}$ (average $u_* = 0.47\ m\ s^{-1}$). By subtracting the contribution from aerodynamic transport, a surface deposition velocity (see paragraph 2.3) was calculated [4]. The surface deposition velocity V_{ds}(loc) is similar in magnitude to V_d(loc) and is 0.00320 ± 0.00231 m s^{-1} on average. The measured V_d(loc) shows only a poor correlation with u_* (Fig. 4). For neutral-unstable conditions the average V_d is 0.00341 ± 0.00196 m s^{-1}, for neutral-stable conditions V_d(loc) is lower, 0.00230 ± 0.00174 m s^{-1}.

The largest uncertainty currently concerns the extrapolation of V_d(loc) determined at 19 m to a V_d representative for the entire canopy. An attempt to derive this scaling factor was recently made Ruijgrok et al. [9], who estimated a value of 2.5. This factor was determined by calculating the collection efficiency for different layers in the canopy, taking into account the leaf area distribution, the activity distribution of ^{214}Pb, the wind profile in the canopy and the different deposition processes involved. Furthermore they assumed that the airborne activity is independent of crown depth. If only impaction is considered, a factor of 2.1 was calculated. Another estimate was obtained by examining the local sulphate flux to branches as a function of crown depth, which yielded a scaling factor of 3.7-3.8.

Applying these scaling factors of 2.1-3.8 to V_d(loc) yields average deposition velocities for ^{214}Pb of 0.0061 - 0.0110 m s^{-1}, with a best estimate of 0.0073 ± 0.0049 m s^{-1}. These values are lower than the deposition velocities found for ammonium(bi)sulphate, which is estimated at 0.012-0.015 m s^{-1} . This is probably related to the difference in size distributions between sulphate aerosol and ^{214}Pb. Apparently deposition mechanisms are less efficient for the smaller particles with attached ^{214}Pb (AMD of 0.4 μm) than for antropogenic sulphate with an MMD of 0.8 μm. A comparison was made between deposition velocities measured for ^{214}Pb and deposition velocities calculated using the parametrizations by Erisman [10] and by Wesely et al. [4]. The parametrization by Erisman yields deposition velocities which are of approximately the same magnitude as the observed values for ^{214}Pb, but the correlation between observed and predicted values is poor. The parametrization by Wesely et al. [4] severely underestimates the deposition velocities.

4. CONCLUSIONS

Deposition velocities for ammonium(bi)sulphate aerosol were derived from vertical concentration gradients measured over a coniferous forest. For different u_*-ranges the median and average deposition velocities were calculated. Deposition velocities range from ≤ 0.004 m s^{-1} for $u_* < 0.20$ m s^{-1} to 0.034 m s^{-1} for u_* between 0.70 and 1.0 m s^{-1}. The median deposition velocity $V_d(50)$ shows a near-linear increase with u_*. Using this relationship the average V_d for the period May - August 1993 is estimated at approximately 0.015 m s^{-1}. This can be considered an upper estimate, due to the possible overestimation of V_d at low turbulence intensity. As a lower estimate a value of 0.012 m s^{-1} is found. It can be concluded that the deposition velocity for sulphate on coniferous forest in The Netherlands has sofar been underestimated by more than a factor two. The annual flux of sulphate aerosol to this site is estimated at approximately 200-240 mol SO_4 ha^{-1} a^{-1}. Measurement of the deposition of ^{214}Pb to branches at the top of the canopy indicates an average V_d(loc) of 0.0029 m s^{-1} m^{-2} leaf area at an average u_* of 0.47 m s^{-1}, but only a poor correlation of V_d(loc) with u_*. Extrapolation of these measurements to the entire canopy results in an average V_d of 0.0061 - 0.0110 m s^{-1}, with a best estimate of 0.0073 m s^{-1}. These values for V_d are a factor 1.6-2.1 smaller than deposition velocities derived from gradient measurements of ammonium(bi)sulphate. The difference is attributed to the smaller particle size of aerosol to which ^{214}Pb is attached.

Acknowledgements
Many colleagues have contributed to this study. We are especially thankfull for the help of Henk Das who formulated the calculational procedures for measurement of low levels of ^{214}Pb activity and of Harry ten Brink who derived an estimate of the fraction of unattached airborne radioactivity. This study was carried out as part of the EUROTRAC-BIATEX programme and the Dutch Priority Programme on Acidification. Funding of the BIATEX-programme by the Ministry of Economic Affairs and the Ministry of Housing, Spatial Planning and Environment is gratefully acknowledged.

REFERENCES

[1] Sehmel, G.A. and Hodgson, W.H. (1980) *AIChE Symposium Series* 76, 218-230.
[2] Hicks, B.B., Wesely, M.L., Durham, J.L. and Brown, M.A. (1982) *Atmospheric Environment* 12, 2899-2903.
[3] Hicks, B.B., Matt, D.R., McMillen, R.T., Womack, J.D., Wesely, M.L., Hart, R.L., Cook, D.R., Lindberg, S.E., De Pena, R.G. and Thomson, D.W. (1989) *J.Geophys.Res.* 94, 13003-13011.
[4] Wesely, M.L., Cook, D.R., Hart, R.L. and Speer, R.E. (1985) *J.Geophys.Res.* 90, 2131
[5] Bosveld, F.C. (1991) Report WR-91-02, KNMI, De Bilt.
[6] Weststrate, H. and Duyzer, J.H. (1994) Report MW-R 94/104, TNO, Delft.
[7] Fowler, D. and Duyzer, J.H. (1989) In: M.O. Andreae and S.D. Schimel (Eds.) *Exchange of trace gases between terrestrial ecosystems and the atmosphere.* John Wiley and Sons Ltd., 189-207.
[8] Van Aalst, R.M. (1986) In: S.D. Lee, T. Schneider, L.D. Grant and P.J. Verkerk (Eds.) *Aerosols.* Lewis Publ., Chelsea MI, 933-949.
[9] Ruijgrok, W., Tieben, H. and Eisinga, P. (1994) Concept report, KEMA, Arnhem.
[10] Erisman, J.W. (1993) *Water, Air & Soil Poll.*, 71(1/2) 81-100.
[11] Khlystov, A., Ten Brink, H.M. and Wyers, G.P. (1993) Report ECN-C--93-011, ECN, Petten.
[12] Porstendörfer, J. (1994) *J.Aerosol Sci.* 25, 219-263.

[13] Bondietti, E.A., Hoffman, F.O.and Larsen, I.L. (1984) *J.Environ.Radioactivity* 1, 5-27.
[14] Schery, S.D. and Wasiolek, P.T. (1993) *J.Geophys.Res.* 98, 22915-22923.