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TNO Division of Technology
for Society

P.O. Box 217
2600 AE Delft
Schoemakerstraat 97
2628 VK Delft, The Netherlands

Telex 38071 zptno nl
Fax +31 15 61 68 12
Phone +31 15 69 69 00

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DRY DEPOSITION OF NH3 OVER FOREST

Author : J.H. Duyzer et al.

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Measurement of the Dry Deposition Flux of NH_3 onto Coniferous Forest

J.H. Duyzer[1], H.LM. Verhagen[1], J.H. Weststrate[1] and F.C. Bosveld[2]

1) TNO Institute of Environmental Sciences, P.O. Box 6011, 2600 JA Delft, The Netherlands
2) Royal Netherlands Meteorological Institute, P.O. Box 201, 3730 AE De Bilt, The Netherlands

ABSTRACT

The dry deposition flux of NH_3 to coniferous forest was determined by the micrometeorological gradient method using a 36 m high tower. Aerodynamic characteristics of the site were studied using a second tower erected in the forest 100 metres away from the first. Fluxes and gradients of heat and momentum measured on both towers indicated a fairly homogeneous turbulent flow field over the studied area of the forest. Site specific flux profile functions for heat were derived from continuous measurements of turbulent fluxes and gradients. These functions were used to derive fluxes from the observed gradients of NH_3. In total eighty 90-minute NH_3 flux runs were performed. The result indicate a strong non-stomatal uptake of NH_3 by the forest. A representative dry deposition velocity for NH_3 of 3.6 cm/s was derived. The annual average flux was roughly estimated to be equivalent to 50 kg N ha/yr, significantly higher than the critical load for coniferous forest.

INTRODUCTION

Dry deposition is the process by which gaseous or particulate material is transported from the atmosphere to the earth's surface without the participitation of precipitation. Dry deposition is studied because it is a removal mechanism for several relevant atmospheric gases and it is at the same time an important pathway by which pollutants from anthropogenic origin enter sensitive ecosystems. NH_3 is an important trace gas because of its potential for acidification after nitrification (van Breemen *et al.*, 1982) and because it may be important as a nitrogen source for ecosystems. Because of intensive animal production in the Netherlands with associated NH_3 emissions, the local NH_3 concentration is relatively large. Yearly arithmetic averages up to 4 $\mu g/m^3$ are reported for some regions (Asman and van Jaarsveld, 1990). On the basis of model calculations it was estimated that

NH_3 contributes for more than 20% to the total load of potential acid into Dutch ecosystems (Schneider *et al.*, 1990). In these calculations the deposition velocity of gaseous NH_3 (by definition the ratio of the flux and the air concentration) is an uncertain parameter.

Few studies on the dry deposition of NH_3 have been reported. These studies are often carried out in the laboratory with crops at high concentrations of NH_3 and the results may not apply in the field (Aneja *et al.*, 1986). Measurements of NH_3 deposition over heathland showed high deposition velocities indicating efficient absorption by the vegetation (Duyzer *et al.*, 1987). Based on these studies it was expected that NH_3 would also be absorbed efficiently by forest as well and lead to high deposition fluxes. In order to decrease the uncertainty in the estimated fluxes an experimental program was set up to determine a representative deposition velocity for NH_3 over forest. In this program measurements of the deposition velocity of NH_3 were carried out in the Speulderbos forest in the centre of Veluwe close to important source regions. On this site several concurrent research projects within the Dutch Acidification Program were in progress while the NH_3 fluxes were being measured.

MEASUREMENTS OF DRY DEPOSITION OVER FOREST

The processes by which air pollutants are transported by eddies from the atmosphere to the vegetation surface are understood relatively well. On the basis of this understanding several methods for measurements of dry deposition fluxes have been developed. Overviews are given by Hicks *et al.* (1978) and Fowler and Duyzer (1989) showing that micrometeorological methods are attractive because they yield directly the relevant quantity i.e. the flux from the atmosphere into the vegetation. Moreover parameterized results from these measurements can directly be applied in air pollution models to estimate deposition fluxes over other larger areas and periods. Most micrometeorological methods are based on the assumption that the vertical flux through a horizontal plane measured at a reference level above the surface provides the flux through the atmosphere/vegetation interface. This assumption is valid when specific conditions regarding stationarity, horizontal uniformity of the absorbing surface and atmospheric conditions, are fulfilled. A principal micrometeorological method is the eddy correlation method where the flux (F) is derived from the covariance of concentration (c) of the entrained quantity and the vertical component of the wind velocity (w) i.e. $\overline{F} = \overline{wc}$.
However, in order to measure the contribution of all eddies to the flux, fast response sensors are necessary. At the moment no equipment is available to measure the NH_3 con-

centration in air sufficiently fast to use the eddy correlation method. In this study the eddy correlation method was used only to measure heat, moisture and momentum fluxes and the gradient method was used to measure the NH_3 flux. Here the flux is derived from measurements of the vertical concentration profile $\left(\frac{dc}{dz}\right)_z$ at height z and a turbulent diffusion coefficient i.e.

$$F = -k_z \cdot \left(\frac{dc}{dz}\right)_z \qquad [1]$$

k_z, the turbulent diffusion coefficient for NH_3 can be derived from measurements of atmospheric turbulence expressed as

$$k_z = \frac{k.u_* z}{\phi_c} \qquad [2]$$

where k = von Karman's constant (0.4), u_* is the so called friction velocity, z is the height over the zero displacement level d and ϕ_c is the dimensionless flux profile relation defined as

$$\phi_c = -\frac{ku_* z}{F}\frac{dc}{dz} \qquad [3]$$

The gradient method has been applied successfully for several trace gases such as SO_2, O_3, NO_x over low vegetation (Duyzer and Bosveld, 1988). A major problem with this method is to measure the vertical gradient which can be very small especially over rough surfaces. Information on the flux profile function (ϕ_c) of trace gases is not normally available. Assuming similarity of transport between trace gases and heat ϕ_c is often calculated from established formulations of these functions for heat (Dyer and Hicks, 1970). Over forest the situation is more complicated. Towers are often a compromise between scientific demands and costs, measurements are often carried out at levels only partly exceeding the roughness layer just above the forest canopy. In this layer substantional deviation from classical flux profile relations may exist (see for example Raupach, 1979). In addition the complexity of forest sites gives rise to advective influences for which especially profiles are sensitive. Because of the lack of a theoretical framework generalization and subsequent use of experimental results from other sites is difficult. Therefore in situ determination of the flux profile functions is necessary.

The modified Bowen ratio method in which the flux is calculated from implicit determination of k_z does not have many of the drawbacks of the gradient method. It necessitates however very precise measurements of air temperature at the same heights as the

concentration measurements and accurate data on sensible heat fluxes (H). The flux is derived as:

$$F = \frac{H}{\Delta\theta} . \Delta C \qquad [4]$$

were $\Delta\theta$ is the potential temperature gradient and ΔC the concentration gradient over the same height.

Over forest under near neutral conditions the temperature gradients may be so small that they cannot be measured with sufficient accuracy. This method therefore has only limited applicability.

Both the Bowen ratio and the gradient method regard the zero displacement level for trace gas as identical to that for heat. This is often not true and therefore contributes a systematic error to the results. Over low vegetation the error is normally small but over forest it may become significant especially for reactive gases such as O_3 or NO that may have additional chemical sinks or sources at different levels to those of heat.

Implementation of micrometeorological methods on the Speulderbos site

In order to increase the accuracy and to decrease the uncertainties related to ammonia flux measurements over the Speulderbos forest, the standard measurement procedure was expanded considerably. The standard experimental set up for heathland described in Duyzer *et al.* (1987) using three denuders at each measurement level was doubled in order to increase the accuracy and to lower the detection limit for flux measurements. This seemed necessary because gradients were expected to be smaller than those over heather. Although this method is very labour intensive it is scientifically attractive because all denuder measurements are in principle completely independent; some systematic errors are therefore avoided. These errors are always hard to overcome with gradient implementations using different monitors or sample tubes for each level.

The presence of a horizontally homogeneous turbulent flux field is the theoretical basis for the extrapolation of the measurements at one tower to the whole forest surface. In order to investigate the turbulent field over the forest a second tower equipped with an identical set of meteorological instruments was erected at some 100 m to the west of the first tower. This tower was operated by the Royal Netherlands Meteorological Institute (KNMI tower). Comparing the measurements of the heat and momentum fluxes at both towers the homogeneity of the flux fields over this area of this forest could be studied. The KNMI tower was also used to perform detailed studies of flux profile functions ϕ for momentum,

heat and water vapour on the site. Profiles of windspeed, air temperature and water vapour concentrations were measured from 36 m (two times the height of the tree tops) down to the canopy level (lower than 18 m). In this way the deviation of normal flux profile relations could be studied and relations specific for this site could be derived.

EXPERIMENTAL

The Speulderbos site is located in the central part of the Netherlands in the middle of an important source region for NH_3 and consists of a relatively small homogeneous area with douglas fir 15 metre in height surrounded by forest of different composition. The forest is dense, the projected leaf area index is 11, typical stem distance is only a few metres. The site is schematically presented in figure 1 (after (Vermetten *et al.*, 1990)). As can be concluded from this figure the forest is not homogeneous over scales of 300 metre and therefore in principle unattractive for meteorological measurements. The surrounding forest however is roughly of the same height. Large transitions in aerodynamic roughness such as heathland/forest occur more than 1500 metres away. Many experiments described here were carried out when the wind was blowing from easterly directions. The NH_3 flux measured at the LUW mast will then also be influenced by the oak forest. The presence of an adequate infrastructure and advantages for carrying out the work close to related research on acidification in the same forest provided the arguments to conduct the present study at this forest. The 30 metre tower operated by the Agricultural University of Wageningen (Department of Air Pollution) for monitoring of gaseous pollutants was used. During measurement campaigns this tower (the LUW tower) was equipped with a comprehensive set of meteorological instruments and extended to 36 metre.

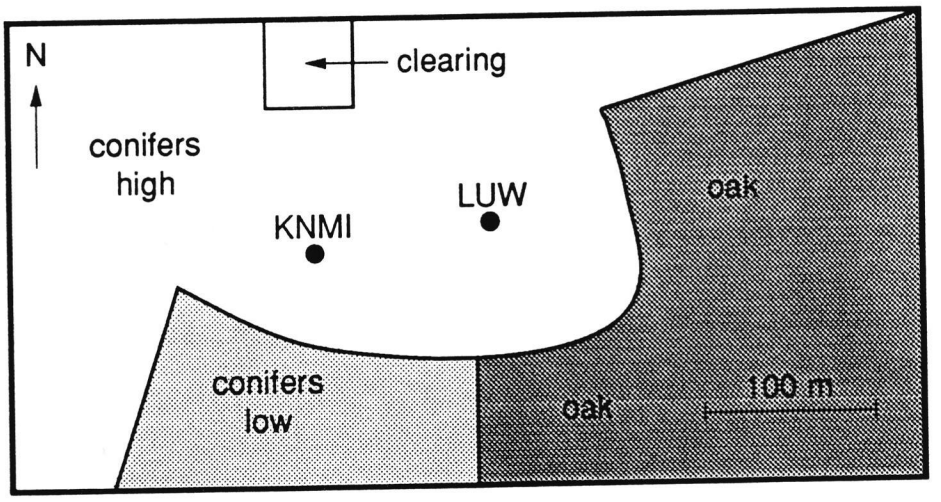

Figure 1 Speulderbos research site.

NH_3 concentration, air temperature and wind velocity were measured at 4 levels i.e. 36, 30, 22 and 18 metres high. Each level consisted of a cup anemometer (A.M. Young) and a ventilated and shielded PT100 thermometer each of these on small booms extended 2 to 3 m from the centre of the mast. Tests with all sensors mounted at one level showed that all temperatures were within 0.015 K. A three dimensional sonic anemometer was mounted on the 30 metre level on a small boom. The boom could be moved in order to position the sonic to provide optimal flow through the sonics centre. Six oxalic acid coated denuders were mounted at each level shortly before each run. Denuders were constructed, prepared and treated according to Ferm (1979). Each denuder was equipped with a calibrated critical orifice and treated separately throughout the whole analytical procedure. As a means of quality control each level was equipped with a mass flow metre system to obtain accurate estimates of sample flow at each level. After 1.5 hour, sampling was stopped and all denuders were dismounted and stored in an NH_3 free environment in order to reduce contamination to a minimum.

After each campaign (typically within 2 weeks after sampling) all denuders were rinsed with a NaOH solution and analysed for NH_4^+ using a coloring agent. The standard error of the average concentration at each level was 0.08 $\mu g/m^3$ (95% confidence level $\frac{2\sigma}{\sqrt{6}}$).

Figure 2 gives a schematic picture of the LUW tower with equipment as mounted during campaigns.

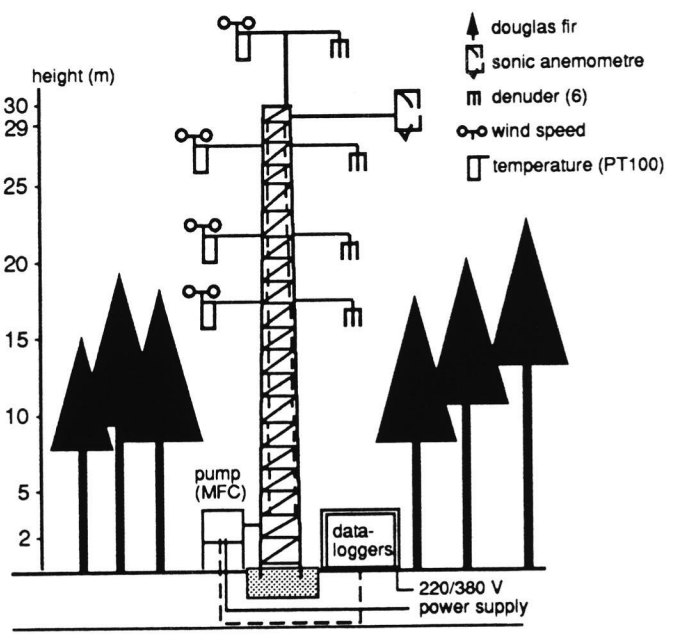

Figure 2 Experimental site with set up for TNO campaign measurements (LUW tower).

On the second tower operated by KNMI a similar set of meteorological equipment was mounted. Several additional meteorological parameters were monitored (rainfall, net and global radiation, effective radiative temperature of the forest canopy and humidity profiles). More detail on the instrumentation in the KNMI mast is given in Bosveld (1990).

RESULTS

From April 1988 to March 1990 six campaigns were carried out in the Speulderbos forest. Campaigns were carried out in all four seasons. During each of these campaigns 10 to 15 independent flux measurements with an averaging time of 1.5 hours were carried out. During the NH_3 sampling runs turbulent quantities such as heat and momentum fluxes and associated profiles were registered on both towers. In 1989 the meteorological measurements on the second tower were carried out continuously in order to obtain statistically reliable flux profile functions. The results of the micrometeorological measurements are presented first. Secondly the results of the NH_3 flux determinations using the modified Bowen ratio method and the gradient method are compared. Finally average fluxes and representative deposition velocities are estimated.

Comparison of eddy-correlation fluxes measured on two towers

A comparison was made between the fluxes for momentum and temperature measured on both towers during campaigns in 1989. Both quantities were measured by identical sonic anemometers at 30 m height. The sonic temperature flux $\overline{wt}$ was used both uncorrected for moisture and wind. The friction velocity was corrected for angle dependence and for upstream round the probe as derived by Duyzer and Bosveld (1987). No correction was made for misalignment of the probe relative to the horizontal plane or the streamlines. Figure 3a en 3b show the relation of the friction velocity and the sonic heat flux at the two locations. Note the high friction velocities up to 1 m/s. In both figures the data are classified according to wind direction. The wind sectors for which it is believed that no mast interference is present are indicated with black. It is seen that for these wind sectors 90-240 the deviations from the 1:1 line are smaller then for the other classes. The friction velocities show a deviation of circa 5%. For the temperature flux the deviation in the undisturbed sectors (the black points) is circa 12%.
In the light of such possible error sources as non-horizontal positioning of the sonics and different obstructing geometry around the sensors the deviations found in the undisturbed sectors are not very large. Therefore it was concluded that this deviation does not present a large contribution to the overall uncertainty of NH_3 flux estimates.

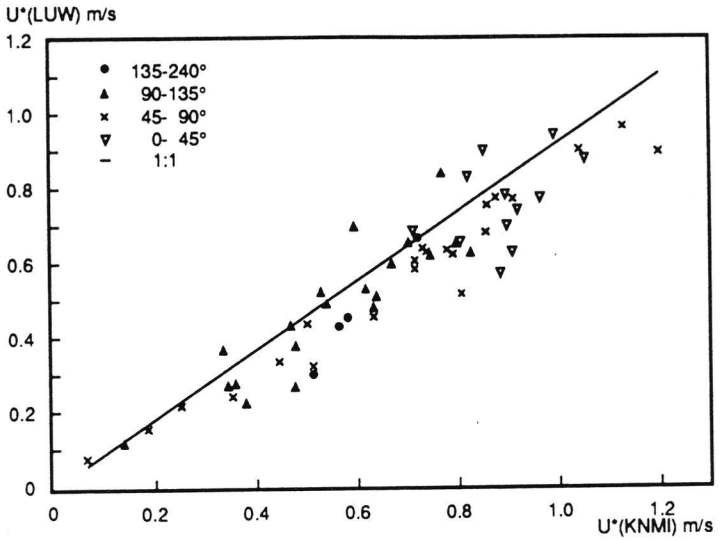

Figure 3a Comparison of u_* values measured by eddy-correlation on LUW and KNMI tower.

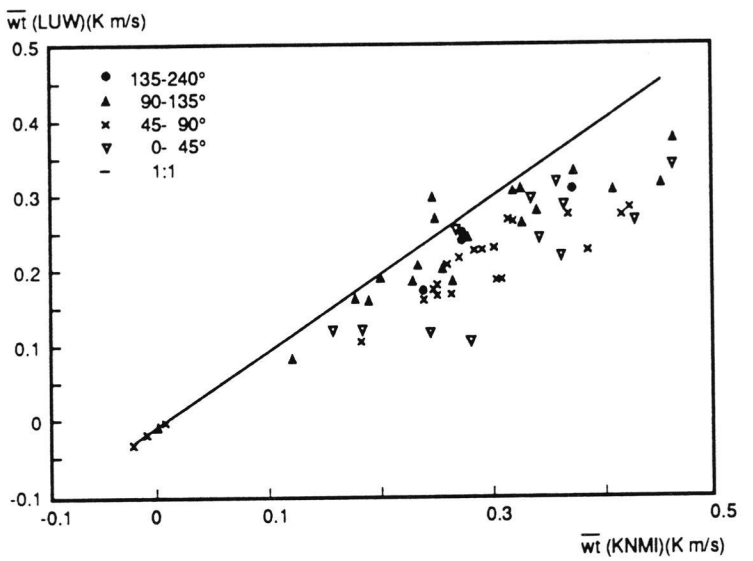

Figure 3b Comparison of heat fluxes measured by eddy-correlation on LUW and KNMI tower.

Estimates of flux profile functions

Figure 4 shows the dimensionless flux profile relations for heat derived from continuous observations at the second mast. Half hour mean values were selected from the period April 1989 to January 1990. Selection of the data was performed to guarantee good instrumental performance and definite de- or ascending temperature profiles. To exclude rapid changing conditions especially during sunrise and sunset only data with absolute heat fluxes larger than roughly 18 W/m^2 (0.015 K m/s) and absolute temperature difference

larger than 0.05 K were used. Data were sorted in order of increasing stability. Ten successive values were used to get one ϕ value by the following procedure. Rewriting equation (1) and (2) for heat transport gives:

$$k.u_* \frac{\Delta\theta}{\Delta \ln(z-d)} = \phi_h\left(\frac{z}{L}\right) . \overline{wt} \qquad (5)$$

where $\Delta\theta$ is the difference between the potential temperatures on the two levels and $\rho c_p \overline{wt}$ is the observed turbulent heat flux (ρ is the density of air and c_p the heat capacity). Potential temperatures were calculated from observed air temperatures T as $\theta_z = T + \gamma z$ where γ is the dry adiabatic lapse rate. ϕ can be calculated now as the linear regression coefficient with zero offset between the left handside equation (4) and $\overline{wt}$. z is calculated as the geometric mean of the two measurement heights.
For each of the three intervals in the profile we arrive at scatter diagrams for ϕ_h. These are displayed in Figure 4a and b.
The ϕ_h value for neutral conditions is smaller than one and decreases with height from 36 to 18 m. The data at the lowest heights show a proportional behaviour while for the stable side the line is displaced parallel. Applying a zero displacement level of 12.5 m for this tower (roughly estimated as $\frac{2}{3}$ of treetop height) the data for all levels can be described quite well with simple modifications of the well known relations given by Dyer and Hicks (1970).

In the roughness layer the following formulations are proposed for the flux profile relations $\phi_{h,z}$

$$L \leq 0 \qquad \phi_{h,z} = \alpha_H \phi_h \qquad \text{where } \phi_h = (1 - 16\frac{z}{L})^{-1/2} \qquad (6)$$

$$L > 0 \qquad \phi_{h,z} = \phi_h - (1-\alpha_H) \qquad \text{where } \phi_h = 1 + 5\frac{z}{L} \qquad (7)$$

L is the Monin Obukhov stability parameter defined by

$$L = \frac{-u_*^3}{k.g.\langle wt\rangle} \quad \text{where g is the acceleration by gravity.} \qquad (8)$$

The correction factors α for flux profile function ϕ_h are given in table 1 and are plotted together with the data points in figure 4a and b.

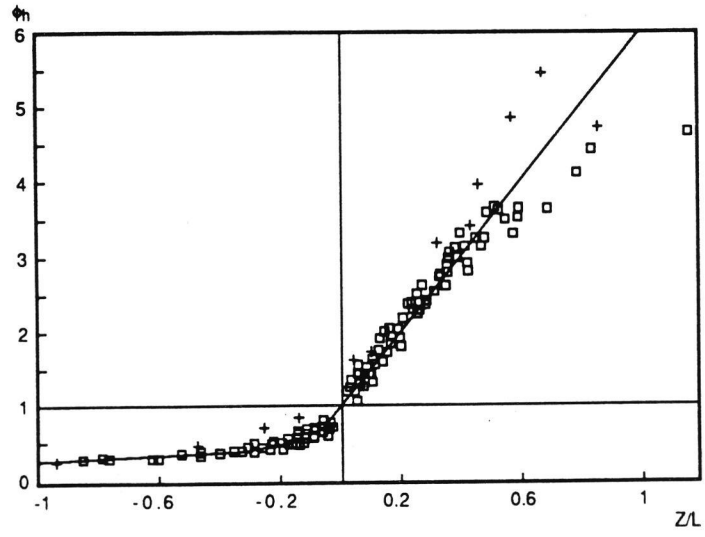

Figure 4a

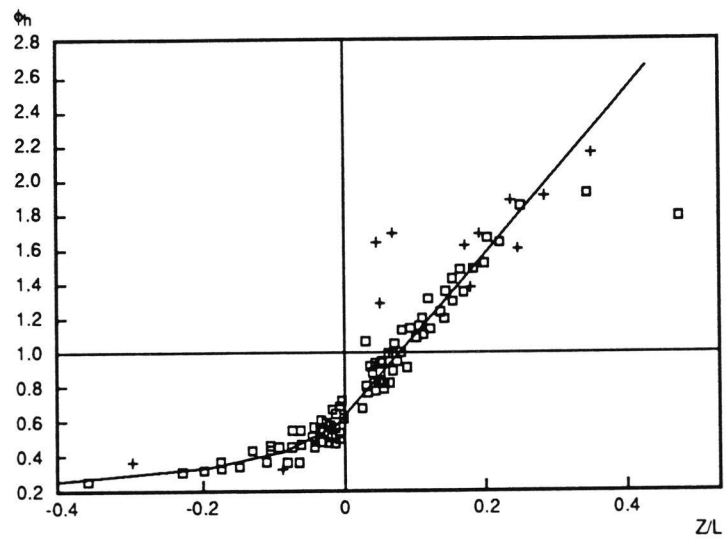

Figure 4b

Figure 4 Dimensionless temperature gradient for two height intervals at the KNMI tower.
a. interval 31-36 m drawn line corresponds to formulas 6 and 7 with $\alpha = 0.95$
b. interval 18-24 m drawn line corresponds to formulas 6 and 7 with $\alpha = 0.65$

Table 1
Correction factor α_H for classical flux profile functions ϕ_h.

Height intervals [m]	α_H
18-24	0.65
24-31	0.80
31-36	0.95

The decrease of α_H approaching the surface can be understood as follows:
While approaching the vegetation the length scale k(z-d) decreases linearly. In the neighbourhood of the vegetation turbulence is generated by the individual trees on length scales related to those of the roughness elements. Relative to its undisturbed value the disturbed turbulent length scale will decrease more slowly than linear. As a result mixing will be more intensive and gradients less steep.
The combination of d, α and the Dyer and Hicks relations is an appropriate mathematical description of the relation between the heatflux and the temperature gradient in this forest. The zero displacement level d is somewhat arbitrarily chosen as $\frac{2}{3}$ of the treetop height. Other choices for d will lead to other estimates for α_H. The physical meaning of d is less important here. See for example Hicks *et al.* (1979) for a discussion on d.
A more detailed description of the measurements and interpretation of the data including those for momentum is given in Bosveld (1990).

Calculation of NH_3 fluxes from gradients

The flux profile functions play an important role in the interpretation of the concentration profiles measured for NH_3. As discussed above the combination of d and ϕ found for heat may not be valid for trace gas. The distribution of leaf area in the canopy shows a maximum around 12 metre. The crown depth is roughly 6 metre (Evers, 1990). It seems reasonable to assume that the maximum absorption of a "sticky" gas such as NH_3 takes place within the crown level. Exact determination of the flux profile functions and the zero displacement level for NH_3 requires independent measurement of the NH_3 flux by, for example, the eddy correlation method.
Measurements of the concentration profile within and below the crown could give some additional information on the zero displacement level for NH_3 relative to that for heat. Since better information is not available it is assumed that these functions are essentially the same as those for heat. The uncertainty introduced by this assumption is probably related to the crown depth and the measurement height and would in this case be from the order of 25%. Therefore:

$$\phi_c = \phi_h = -\frac{k.u_* z}{F}\frac{dc}{dz} \tag{9}$$

integration leads to

$$c_z = -\frac{F}{ku_*}\left[\ln\frac{z}{z_0} - \left(\psi_h\left(\frac{z}{L}\right) - \psi_h\left(\frac{z_0}{L}\right)\right)\right] \tag{10}$$

where

$$\psi\left(\frac{z}{L}\right) = \int_{o}^{z} \frac{1-\phi_h\left(\frac{z}{L}\right)}{z}\, dz \qquad (11)$$

The integrated forms of the profile functions are well known normally and formula (10) can be used to derive the flux. An attractive way to do this is by calculating the slope F/ku_* of the line described by the plot of c_z against $x=\ln(z) - \psi\left(\frac{z}{L}\right)$. The advantage of this method is that the standard statistics of the straight line can be used to judge the quality of the observations. For instance it is possible to judge whether a flux is significantly different from zero and to trace outliers in the NH_3 measurements that are outside the 95% confidence interval of the line. The relative error in the slope as can be calculated using a least square method is equal to:

$$r.e = \sqrt{\frac{1-1/\rho^2}{N-2}} \qquad (12)$$

where ρ is the correlation coefficient between c and x and N the number of observations. The scatter in the concentration measurements is usually so large that it dominates the scatter in the flux estimate. Typical standard deviation in 9 consecutive 10 minute averages of u_* is 10-20%. For NH_3 flux estimates an averaging period of 90 minutes was used. Over this period the 95% confidence interval of u_* is less than 10%. The statistical error in the estimate of the slope dc/dlnz is usually larger and thus dominates the error in the flux.
In this case over forest the calculation is not as straightforward because ϕ_h is not a simple function of height, its value is only known at a few points and thus integration to obtain ψ is not simple. Assuming for practical purposes that the classical flux profile functions are valid at the 36 m level the integrated values and x values for the regression of c_z as a function of x(i) were derived from:

at 36 m (i=4) $x(i) = \ln z_i - \psi_h\left(\frac{z_i}{L}\right)$ where $\psi_h\left(\frac{z}{L}\right)$ is the classical integrated flux profile function. At the other levels x values for the least square calculations are derived from

$$x(i) = x(i+1) - \left(\log\left(\frac{z_{i+1}}{z_i}\right) - \psi_h\left(\frac{z_{i+1}}{L}\right) + \psi_h\left(\frac{z_i}{L}\right)\right) \cdot \alpha_i \qquad (13)$$

for unstable conditions and

$$x(i) = x(i+1) - \left(\log\left(\frac{z_{i+1}}{z_i}\right).\ \alpha_i - \psi_h\left(\frac{z_{i+1}}{L}\right) + \psi_h\left(\frac{z_i}{L}\right)\right) \qquad (14)$$

for stable conditions.

These formulations can be obtained from rewriting equations (6) and (7). As a result of this transformation a plot of c_z against x_z yields a straight line with slope equal to $\frac{-F}{ku_*}$. Using the average of 9 consecutive 10 minute values for u_* and H (to calculate L) the flux of NH_3 and its relative error can be determined.

NH_3 fluxes

In a first screening some 10 measurements were removed from the dataset because the standard analytical quality was not achieved. This judgement was based upon the standard deviation of the six independent concentration measurements at each level which is normally some 5% of the mean value. Measurements were rejected when this standard deviation at more than one level was larger than 20%.

Based on normal student T test, gradients were not considered to differ significantly from zero when the relative error in the slope was larger than 46%. Out of a total of some 80 complete measurement runs 50 showed smaller errors with a median value of some 25%. In the rest of the dataset errors were often over 100%. This means that although the analytical measurements are of good quality it is statistically not certain whether the flux is upward or downward. Still the result is valuable because the flux is certainly between two limits (for example 95% confidence intervals). Therefore these data have to be used to calculate the average flux of a limited dataset. Using them to calculate the deposition velocity will contribute a large unrealistic scatter to the data only. The good quality data can be used to study the influence of process-level parameters such as windspeed, temperature etc. Details of the 50 satisfactory measurements are presented in the appendix.

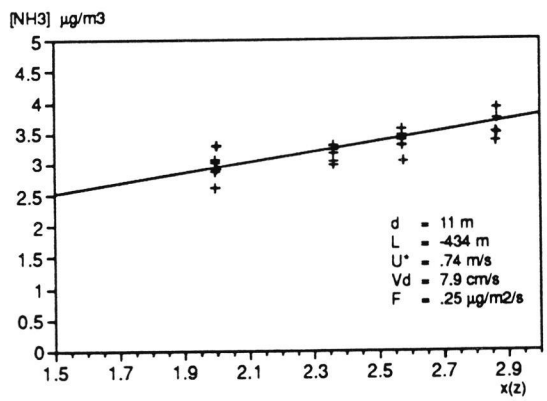

Figure 5 Results of denuder measurements at 4 levels plotted as a function of x(z) (see text). (16 November 1989 12:06-13:36)

An example of an observed gradient is depicted in figure 5. It is obvious from this graph that the gradients over the forest are small compared to the scatter in the denuder measurements. The difference between the concentration at the highest and the lowest level is roughly 20%. The deposition velocity is close to the maximum possible, limited only by the turbulent transport rate to the canopy.

In order to test the validity of the use of the corrected flux profile functions the flux was also calculated using the modified Bowen ratio approach. From 4 measurement heights 6 combinations can be made and an average value can be calculated according to:

$$\bar{F} = \frac{1}{6} \sum_{i=2,4\ j=1,3} \frac{H}{\overline{\theta_t} - \overline{\theta_j}} \cdot (\overline{c_i} - \overline{c_j}) \qquad (15)$$

where i and j are different measurement levels. These values are now compared to the flux estimates based upon the approach described above i.e. the NH_3 flux is calculated from the slope of the line through all 24 denuder measurements plotted against the x(i) values according to equation (13). The result of this comparison is shown in figure 6. The graph shows fluxes towards as well as fluxes away from the surface (positive sign). Apart from a few outliers fluxes calculated according to both methods are in good agreement over the whole range with no systematic differences. This result supports the choice that was made for the zero displacement level and the integrated flux profile functions.

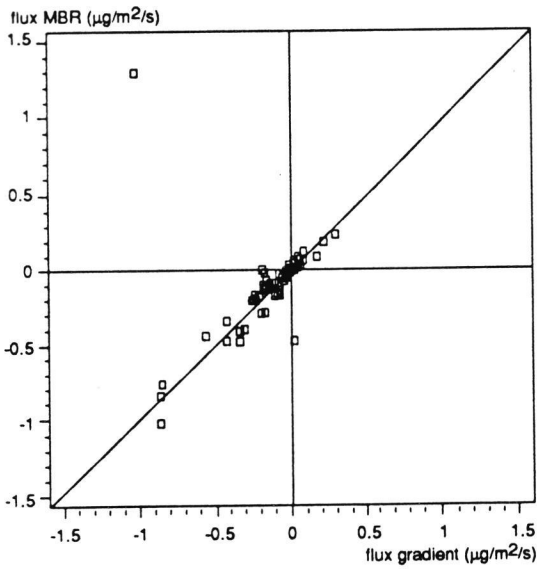

Figure 6 NH_3 flux calculated according to the modified Bowen ratio (MBR) method and the gradient method. Positive signs indicate emission fluxes. The outlier at the left hand corner is an example of a case with neutral stability where the Bowen ratio method is less applicable.

The relation between the flux and the concentration is shown in figure 7. Fluxes not significantly different from zero are indicated different from the others. For illustrative purposes only a line corresponding to a deposition velocity of 5 cm/s is drawn. This line seems to give a reasonable agreement with the "good quality" measurements. Most of these data give fluxes towards the surface. Many of the observed emission fluxes have large errors and the direction of the flux is not certain. At a few occasions however, most at concentration levels around 1 $\mu g/m^3$ good quality measurements of emission fluxes are found. There may be several explanations for these phenomena such as evaporation of NH_3 from a drying surface or a compensation point for NH_3. Advection fluxes are not very likely because they are probably coupled to high concentrations. The complete dataset gives an average v_d of 2.2 (σ = 5 cm/s). The average flux over all measurements is equivalent to roughly 40 kg N/ha/year.

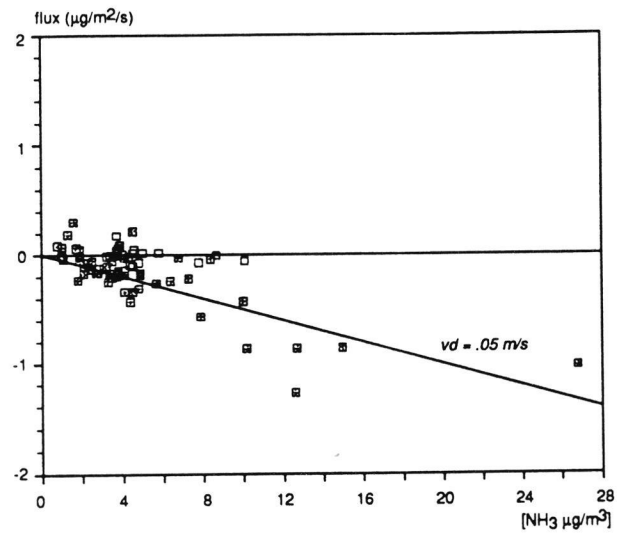

Figure 7 Flux of NH_3 plotted against NH_3 concentration. Positive sign indicates emission. open squares indicate fluxes with an error larger than 46%.

Generalisation of the results

Very often the deposition process is parameterized using the resistance analogy. This is done to separate the influence of the meteorological parameters from the parameters that are determined by properties of the vegetation surface. By doing this the variability in the results caused by atmospheric variability is reduced. Moreover it provides the link between deposition and the controlling physical, chemical or biological processes. More detail about the surface layer model is given for instance in Hicks and Matt (1988).

The canopy or surface resistance r_c the parameter that describes the actual uptake process at the surface is calculated from measurements of v_d as $r_c = \frac{1}{v_d} - r_a - r_b$.

r_a is the resistance to transport of the turbulent layer of air whereas r_b is related to the resistance to transport to the still layer of air surrounding the vegetation elements.
For O_3 and SO_2 the model works quite well because r_a and r_b are usually small compared to r_c and from measurements it was concluded that uptake of both gases by many plants is via stomata. This is a very important result for generalization because much information about stomatal dynamics is available from previous studies on water vapour exchange. For forests r_b is very important when r_c tends to zero. The existing understanding of r_b however is poorly developed for trace gas exchange especially over rough surfaces. This uncertainty is reflected in the uncertainty in r_c calculated from the measurements for extrapolation to other sites.
The calculated resistances for each run are given in the appendix. Typical daytime aerodynamic resistances vary between 10 and 20 s/m. The average surface resistance is close to zero with a standard deviation of 30 s/m. In a few cases negative surface resistances are calculated. These are physically impossible but are probably related to statistical errors and uncertainties in the formulation of the surface layer model.
Although the scatter in the results is quite large it is obvious from our measurements that the surface resistance of coniferous trees is low (median and average value around 4 s/m). This value is much lower than the stomatal resistance for water vapour (r_s) which was found to be larger than 100 s/m for this forest (Bosveld *et al.*, 1991). The uptake rate of NH_3 by stomata is therefore limited to $1/r_s$ i.e. a maximum deposition velocity of 1 cm/s. Deposition velocities of NH_3 of over 4 cm/s (with close to zero r_c) were observed, even in cases with a clearly dry forest. These observations give evidence of an additional uptake much stronger than the stomatal uptake. Only in a few cases high surface resistances were found. Most of these were coupled with windspeeds lower than 2 m/s when the use of the gradient approach and the resistance layer model is less valid. The results seem to be similar to those obtained by Duyzer *et al.* (1987) over heathland where a surface resistance of 43 s/m was found during dry conditions and 12 s/m was found during wet conditions. The lower surface resistance found over the forest in dry conditions may be related to the large leaf area index of the canopy. The results of the measurements reported here support the idea of strong absorption under wet conditions but the difference with dry conditions is not significant because of the limited number of measurements carried out under wet conditions.
Only two successful measurements were carried out during the night. The deposition fluxes were small compared to daytime fluxes probably because of the increased resistance to transport of the boundary layer. The fluxes were equivalent to less than 10 kg N/ha with a deposition velocity of less than 0.4 m/s. The interpretation of these measurements is

troubled by instationarity resulting from the developement of a strong inversion. Therefore generalization of the night time results is impossible. On the other hand there is at this stage no reason to assume that the (non stomatal) uptake during the night is different from day time uptake. Moreover the probability that the canopy is wet at night is high. Therefore it is assumed here that the surface resistance for NH_3 at night is equal to the day time values i.e. practically zero.

An average diurnal deposition velocity

Based on our measurements it is tentatively concluded that the surface resistance to NH_3 uptake is low throughout the year. To estimate a yearly average dry deposition velocity it is essential to take all meteorological conditions through the year into account, especially the influence of night-time stability.
This was done using the resistance layer model by calculating the aerodynamic resistances r_a and r_b as:

$$r_a = \frac{U_z}{u_*^2} \tag{16}$$

$$r_b = \frac{2}{ku_*} (Sc/Pr)^{2/3} \tag{17}$$

where U_z = wind speed at height z. Sc and Pr are the Schmidt number and the Prandtl number respectively.
For every half hour in 1989 r_a and r_b were calculated from the turbulence measurements on the KNMI tower according to (16) and (17) and combined with $r_c = 4$ s/m according to $v_d = \frac{1}{r_a+r_b+r_c}$. The deposition velocities obtained for each half hour averaged over the year are shown in figure 8. The maximum deposition velocities calculated for midday are quite close to the experimental results for this period of the day (median v_d of good quality measurements is between 4 and 5 cm/s). This indicates that our campaign measurements are a reasonable sample of daytime meteorological conditions. It is also interesting to see the strong influence of the night-time stability suppressing the exchange of NH_3 between atmosphere and canopy.

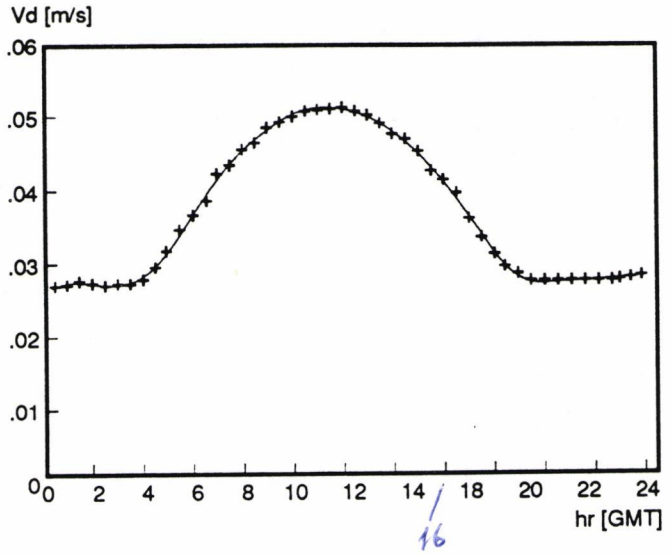

Figure 8 Diurnal cycle of the average dry deposition velocity of NH_3 to coniferous forest based upon atmospherical resistances calculated from actual measurements and r_c = 4 s/m.

The 24 hour average deposition velocity for the year using this approach is 3.6 cm/s. The average v_d is much higher than the value observed over heathland (1.6 cm/s) in earlier studies (Duyzer *et al.*, 1987). This is partly due to the lower surface resistance of forest and partly to the low aerodynamic resistances caused by the roughness of the forest.
Combined with the average NH_3 concentration of roughly 5 $\mu g/m^3$ reported for the site by Vermetten *et al.* (1990) this leads to a dry deposition flux of 50 kg N/ha/yr. A better estimate could be obtained when actual half hour average NH_3 concentrations are combined with estimated deposition velocities. In that way the bias caused by correlations between NH_3 concentrations and deposition velocities can be precluded.
The uncertainty in the present estimate is not easy to assess.
Campaigns were carried out in all seasons and periods were not selected for any special condition. The average in time however is still limited. Compensation for this aspect with respect to meteorological conditions is carried out using the resistance analogy.
The surface resistance r_c is taken very low; using a lower value hardly leads to higher fluxes because the resistance to transport mainly lies in the air compartiment. The surface resistances that were found all show that the forest acts as a perfect sink for NH_3. In some periods of the year temporary saturation of the surface could occur, leading to an increase in surface resistance or even small emission fluxes. Significant emission fluxes were found in a few cases most of them at concentrations lower than 2 $\mu g/m^3$. This could be an indication of a compensation point for NH_3. Since on this site concentrations higher than 2 $\mu g/m^3$ are not often observed (Vermetten *et al.*, 1990) this has only a small effect on the

annual averages. However one should be careful to extrapolate these results to more remote sites with low NH_3 concentration.
In any case this input to this forest exceeds the critical load level for coniferous forest which was estimated to be from the order 10-20 kg of N in total (Nilsson, 1987).

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APPENDIX Details of good quality NH_3 flux estimates

Date	Time	u m/s	u* m/s	L m	T °C	r_a s/m	r_b s/m	NH_3 µg/m³	Flux µg/m²/s	Sigma	V_d m/s	r_C s/m	wet-ness
7 Apr 88	18:15	3.6	0.66	-1744	14.44	8.1	7.1	26.8	-1.03	0.23	0.039	10.2	0
28 Apr 88	13:45	3.0	0.67	-101	10.26	6.8	7.0	10.0	-0.43	0.26	0.044	9.2	0
28 Apr 88	16:05	2.8	0.69	-99	11.8	6.0	6.9	12.7	-0.87	0.21	0.069	1.6	0
29 Apr 88	13:16	2.7	0.48	-39	10.13	11.6	9.8	1.6	0.29	0.06	(-0.181)	-	1
11 May 88	13:39	2.9	0.81	-113	18.95	4.4	5.8	4.1	-0.35	0.10	0.086	1.5	0
7 Sep 88	16:49	2.7	0.63	-179	20.19	6.7	7.5	12.6	-1.27	0.18	0.104	-4.5	0
8 Sep 88	16:50	2.9	0.64	-285	20.23	7.0	7.4	4.4	-0.43	0.39	0.103	-4.6	0
21 Sep 88	11:19	1.0	0.17	-10	12.93	34.9	27.8	15.0	-0.86	0.16	0.058	-45.4	0
22 Sep 88	11:13	2.9	0.61	-368	13.18	7.8	7.8	7.9	-0.57	0.23	0.074	-2.0	0
22 Sep 88	12:49	2.5	0.50	-102	16.5	10.2	9.4	4.8	-0.31	0.38	0.067	-4.5	0
22 Sep 88	13:40	3.2	0.51	-153	17.86	12.2	9.2	10.2	-0.87	0.24	0.087	-9.8	0
25 May 89	12:10	2.2	0.43	-24	24.63	12.2	11.0	4.5	0.21	0.39	(-0.047)	-	0
26 May 89	11:40	2.7	0.66	-77	17.84	6.1	7.2	4.1	-0.19	0.22	0.047	8.0	1
26 May 89	13:52	3.7	0.73	-93	20.0	7.0	6.4	5.7	-0.27	0.21	0.047	7.9	0
26 May 89	15:50	4.0	0.83	-159	19.75	5.9	5.7	6.4	-0.25	0.27	0.039	14.0	0
13 Jun 89	14:52	3.0	0.64	-92	22.83	7.4	7.4	3.2	-0.11	0.23	0.035	13.8	0
13 Jun 89	17:01	2.9	0.58	-67	23.47	8.7	8.2	3.5	-0.06	0.33	0.018	38.3	0
13 Jun 89	23:51	1.9	0.19	20	17.56	52.6	24.8	9.5	-0.03	0.17	0.004	196.5	
14 Jun 89	13:52	1.8	0.40	-17	24.06	11.7	11.9	1.3	0.18	0.12	(-0.133)	-	0
14 Jun 89	15:51	1.7	0.30	-10	24.74	18.7	15.8	1.0	0.06	0.29	(-0.066)	-	0
3 Jul 89	11:20	3.7	0.73	-142	19.66	7.0	6.4	2.1	-0.18	0.19	0.089	-2.1	1
3 Jul 89	12:30	3.2	0.62	-146	20.3	8.1	7.6	2.1	-0.15	0.19	0.075	-2.4	0
3 Jul 89	14:30	3.5	0.62	-117	21.2	9.3	7.6	1.9	-0.06	0.24	0.032	14.5	0
3 Jul 89	16:40	3.2	0.66	-192	21.32	7.4	7.1	2.5	-0.06	0.31	0.025	25.8	0
4 Jul 89	11:42	3.8	0.76	-176	18.81	6.7	6.2	2.9	-0.13	0.16	0.045	9.3	0
4 Jul 89	14:02	4.6	0.90	-206	20.91	5.6	5.2	3.5	-0.22	0.12	0.063	4.9	0
27 Oct 89	14:02	2.5	0.51	-4728	15.69	9.5	9.2	4.9	-0.20	0.25	0.041	5.8	1
7 Nov 89	16:22	1.8	0.29	318	7.79	21.1	16.0	4.9	-0.18	0.09	0.039	-11.5	1
16 Nov 89	13:40	3.3	0.74	-434	5.97	6.1	6.4	3.3	-0.25	0.15	0.079	0.3	0
16 Nov 89	15:50	2.7	0.63	4156	6.35	6.7	7.4	3.8	-0.17	0.22	0.046	7.6	0
17 Nov 89	11:21	3.4	0.52	-758	2.75	12.9	9.2	1.1	-0.04	0.25	0.043	1.5	0
17 Nov 89	13:30	3.7	0.82	-495	5.0	5.5	5.8	1.8	-0.24	0.14	0.013	-3.6	0
17 Nov 89	15:31	3.5	0.78	-2160	5.3	5.7	6.0	2.8	-0.17	0.32	0.062	4.5	0
21 Nov 89	14:11	1.3	0.24	4787	10.55	21.9	19.5	6.8	-0.03	0.35	0.005	164.8	1
21 Nov 89	16:31	1.3	0.17	28	8.87	45.9	28.4	8.4	-0.05	0.05	0.006	91.4	1
21 Mar 90	17:20	5.1	0.97	-582	13.08	5.5	4.9	7.3	-0.22	0.40	0.030	22.6	0
22 Mar 90	14:00	3.6	0.76	-134	11.1	6.4	6.2	4.5	-0.34	0.24	0.078	0.3	0
22 Mar 90	17:50	2.9	0.74	-1553	9.54	5.3	6.4	3.4	-0.17	0.28	0.052	7.6	0
27 Mar 90	15:40	2.2	0.52	-67	7.4	8.1	9.1	3.8	-0.18	0.15	0.049	3.2	0
28 Mar 90	11:10	1.8	0.40	-42	5.43	10.9	11.8	2.3	-0.08	0.32	0.035	5.6	0
28 Mar 90	13:10	3.1	0.63	-130	7.9	7.8	7.5	2.5	-0.14	0.24	0.056	2.4	0
28 Mar 90	15:20	2.7	0.59	-200	8.42	7.8	8.0	2.7	-0.17	0.11	0.063	0.0	0
28 Mar 90	17:40	2.8	0.63	-556	8.08	7.0	7.5	3.7	-0.20	0.15	0.055	3.7	0
29 Mar 90	10:50	1.9	0.46	-77	5.95	9.0	10.3	1.7	0.06	0.38	(-0.036)	-	0
29 Mar 90	16:50	2.3	0.60	-82	9.82	6.3	7.8	0.8	0.08	0.18	(-0.101)	-	0

u = windspeed at 22 m level
u* = friction velocity (sonic anemometer)
L = monin obukhov length scale
T = temperature at 22 m level
r_a = aerodynamic resistance at 22 m level
r_b = laminar sublayer resistance
NH_3 = average ammonia concentration from all levels
Flux = ammonia flux estimated from gradient method
sigma = relative standard deviation in the slope according to (12) (fraction)
V_d = deposition velocity at 22 m
r_c = surface resistance (s/m)
wetness = observed canopy wetness 1 wet, 0 dry