CHAPTER 7 THREE CASE STUDIES

Introduction

In this chapter three experiments will be described which were aimed at determining deposition parameters for different receptors, a heathland, a Douglas fir forest and complex terrain, to be used for generalisation. The first study id the Elspeetsche Veld experiment, conducted at a heathland in the centre of The Netherlands. Micrometeorological measurements of SO_2, NH_3 and NO_2 were made together with throughfall measurements of SO_4^{2-}, NO_3^- and NH_4^+ to determine the annual input to the heathland, to derive deposition parameters and the interrelations between parameters for different gases to be used for generalisation (see Chapter 4) and to study the relation between atmospheric deposition and the soil load (throughfall). The second field study deals with a large scale measurement program in the forested area the Utrechtse Heuvelrug in the centre of the Netherlands to determine the influence of canopy structure and complex terrain (forest edges and roughness transitions) to the deposition of acidifying components and base cations, using the throughfall method. The third field experiment described in this chapter has been conducted in the Speulder forest in the middle of the Veluwe, the largest forested area in the centre of The Netherlands. Micrometeorological measurements were combined with concentration measurements and meteorological measurements to determine the exposure to and input of gases and particles to the forest. Also throughfall measurement were made to determine the soil load and to relate the soil load to the atmospheric load. At this site also long-term measurements of changes in effect parameters were made, together with manipulation experiments. The results of these experiments and those of the inputs are used to evaluate dose - effect relations and to determine the most serious impact at the forest.

7.1 THE ELSPEETSCHE VELD EXPERIMENT ON SURFACE EXCHANGE OF TRACE GASES

7.1.1 INTRODUCTION

A three-year experiment was conducted at the Elspeetsche Veld heathland in the Netherlands (Veluwe). The aim of this experiment was to quantify atmospheric fluxes and to derive deposition parameters for heathland vegetation to use for generalisation. Gradients of SO_2 (RIVM), NH_3 (KEMA and ECN) and NO_2 (TNO) have been measured together with throughfall and bulk precipitation fluxes (RUU). A second aim of this study was to investigate the co-deposition of NH_3 and SO_2 and the relation between throughfall and atmospheric deposition for acidifying components.

In this chapter a summary of the results of this integrated study is given. Detailed descriptions of the measuring methods, equipment and results can be found in the reports and papers published by the individual research groups, i.e. for NH_3 by van den Beld and Römer (1990) and by Wyers *et al.* (1992, 1993), Erisman and Wyers (1993), for SO_2 by Erisman *et al.* (1993), Erisman (1992), Mennen *et al.* (1992) and by Erisman and Duyzer (1991), for NO_2 by Duyzer *et al.* (1991) and for the throughfall measurements by Bobbink *et al.* (1990; 1992). For an extensive description of the experiments the reader is referred to Erisman et. al. (1991) and Erisman (1992).

7.1.2 STUDY AREA AND METHODS

Study area

The experiment was carried out at a heathland nature reserve in the central part of the Netherlands: Elspeetsche Veld (52° 16' N, 5° 45' E), near the village Elspeet. The heathland is located in a region with moderate ambient concentrations of SO_2 (6 $\mu g\ m^{-3}$) and NOx(20 ppb) (RIVM, 1989). The ammonia emission density in this area is about equal to the average NH_3 emission in the Netherlands (Erisman, 1989). The vegetation at the site belongs to the GENISTO-CALLENETUM (dry inland heath, characterised by Calluna (Heather)). At the site Calluna vulgaris (L.) Hull completely dominates the dwarf-shrub layer. Approximately 20% of the soil surface is not covered by vegetation. The age of the Calluna plants is 4 - 5 years and the height of the canopy is 20 - 30 cm (Bobbink *et al.*, 1990). Subsoil is nutrient-poor fluvio-glacial sand with a well developed podzolic soil profile.

The heathland vegetation is surrounded by agricultural grasslands with livestock farms in the South and West at ≥ 1 km distance. The ideal fetch to fulfil the demands of steady state homogeneous flow over the heathland is obtained at about 120-270°. In all other directions the fetch is disturbed by roughness elements too close to the measuring site.

A small part of the field was enclosed for the equipment. Just outside the fence three meteorological towers were erected, one for the NO_2 gradients (during two months), one for the NH_3 gradients using the continuous flow denuders (during three months) and the third for the SO_2 and NH_3 (thermodenuder) gradients. The monitors for SO_2 and NO_2 were stored together with the other sensors and data acquisition equipment in special low boxes within 2 m North of the towers. The NH_3 thermodenuders were stored in a caravan at 15 m North of the towers. Three open rain samplers were placed at 40 cm above the soil surface within the fence. Throughfall and stemflow devices were placed randomly at 5 places outside the fence within a radius of 50 m. In the next section a brief overview of the equipment is presented. Details are given in the references mentioned earlier.

Measurement methods and approach
In Table 7.1, an overview is presented of the different methods and equipment used during the experiment. In order to monitor dry deposition fluxes on a routine basis, dry deposition monitoring systems for SO_2, NO_2 and NH_3 have been developed based on the micrometeorological gradient technique. The SO_2 system was previously tested during a two year feasibility at Zegveld, a grassland location in the centre of the Netherlands (Erisman *et al.*, 1993). Selection criteria have been derived to select measuring periods fulfilling the demands of the flux-profile theory. Furthermore, a scheme has been developed to calculate yearly average fluxes from the selected and rejected measuring periods. After the first year of measurements at Elspeet, the system was considerably improved by using more accurate SO_2 monitors, with a lower detection limit.

TABLE 7.1 Overview of methods and equipment used during the Elspeetsche Veld experiment.

Method	Period	Flux averaging time	Meteorological measurements	Gas analysers	Reference
SO_2 gradients (4, 2, 1 and 0.5 m)	1-4-1989 28-7-1992	30 minute	u, θ, σ_θ, at 5 m; net radiation; 1.5m T and rh at 2.5m height	Thermo Environmental Instruments model 43S	Haan (1988), Mennen *et al.* (1992; 1993) Erisman *et al.*, (1993)
NH_3 gradients (4 and 1 m)	1-4-1989 1-4-1990	30 minute	(meteo of SO_2 gradients used)	thermodenuder	van den Beld and Römer (1990)
NH_3 gradients (4, 2 and 1 m)	6-9-1991 6-12-1991	6 minute	(meteo of SO_2 gradients used)	continuous flow denuders	Wyers *et al.* (1992; 1993)
NO_2 gradients (4, 2, 1 and 0.5 m)	1-10-1989 24-12-1989	1 minute	sonic anemometer	Scintrex LMA3 luminol NO_2 monitor	Duyzer *et al.* (1991)
throughfall, stemflow and bulk precipitation	27-4-1989 27-4-1990	bi-weekly		polythene funnels ($165\ cm^2$)	Bobbink *et al.*, (1990); Bobbink *et al.*, (1992)

7.1.3 CALCULATION OF DEPOSITION PARAMETERS

The standard deviation of the wind direction σ_θ, the wind speed u, temperature T and net radiation R_n are used to estimate the friction velocity u_* and the sensible heat flux (Hicks *et al.*, 1987, Erisman *et al.*, 1993). During a period of six weeks, simultaneous measurements using an automated eddy correlation system were made at this site. u_* values calculated from measurements by the monitoring system agreed very well with the directly measured u_* values (Erisman and Duyzer, 1991). The average stability-corrected concentration gradients of SO_2, NO_2 and NH_3 are calculated as the slope of the regression line of the concentrations as a function of the stability-corrected logarithm of the height (see e.g. Erisman *et al.*, 1993). The flux F, deposition velocity V_d, aerodynamic resistance R_a, quasi-laminar layer resistance R_b and the surface resistance R_c are calculated as 2 hour averages according to the equations given e.g. in Hicks *et al.*, 1987 and Erisman *et al.* (1993).

SO_2

For measurements of the SO_2 dry deposition flux, two periods can be distinguished, 1989 to 1991 and 1991 to half 1992. During 1989 to 1991, measurements were made with the 'old' monitors. Results for this period are given in Erisman *et al.* (1993). These results are summarised in the tables below. For the interpretation of the measurements from the period 1991 to 1992, using the more accurate monitors, rejection criteria have been applied to assure high quality results. These criteria differ somewhat from those given in Erisman *et al.* (1993). The criteria for acceptance are listed in Table 7.2.

TABLE 7.2 Criteria for acceptance of observations and no. of observations rejected (total no. of measurements: 5298 two hourly average periods).

Criteria	'New' monitors	Number of remaining periods after application of criteria
Flux profile relation valid; wind speed at 10 m height:	> 1 m s^{-1}	1031
Non-disturbing fetch; wind direction between:	120° and 270°	1936
Concentration (detection limit)	> 1 µg m^{-3}	604
Concentration difference of two monitors measuring simultaneous at 4 m level:	$\Delta c < 3$ µg m^{-3}	125
$\lvert V_d \rvert < 2\,[R_a + R_b]^{-1}$	$\lvert V_d \rvert < 2*[R_a + R_b]^{-1}$	50
Error in R_c:	< 1000 s m^{-1}	161

Deposition parameters for selected periods

The rejected data in 1991 and 1992 contain 74% of the total number of measurements, 10% less than the measurements during the first period using the 'old' monitors. The average SO_2 concentration at 4 m height for this period was 6.9 µg m^{-3} (ranging from 0.1 to 120 µg m^{-3}). The average SO_2 concentration for the selected dataset was 7.2 µg m^{-3} (ranging from 0.5 to

120 μg m^{-3}). The yearly average z_0 value for the wind direction sectors used in the selected dataset is about 4 cm. This value agrees with those obtained from the eddy correlation measurements (Erisman and Duyzer, 1991). The average R_c values for dry and wet conditions and for daytime and night time are listed in Table 7.3. In this table uncertainty estimates are given based on error propagation calculations using uncertainties in measured quantities (Erisman *et al.*, 1990; 1993; Erisman and Duyzer, 1991; Mennen *et al.*, 1992). The uncertainty in R_c values is given here as if R_c is normally distributed. This is not the case. However, uncertainties should be interpreted as a measure of the accuracy of the mean.

TABLE 7.3 Average SO_2 R_c (4 m) values (± uncertainty estimates) for selected daytime and night-time and dry and wet conditions at Elspeetsche Veld.

Condition	R_c (s m^{-1}) day	R_c (s m^{-1}) night
	old monitors (1989/1991)	
Dry	55 ± 115	95 ± 95
Wet	10 ± 15	25 ± 25
	new monitors (1991/1992)	
Dry	150 ± 245	85 ± 130
Wet	35 ± 35	15 ± 25

R_c values show strong variations with time; when the surface becomes wet, R_c values drop to zero, whereas at very dry conditions R_c values can easily increase up to values of 10000 s m^{-1} at night. By the use of average R_c values for only four different classes, this large variation with time is not accounted for. An R_c parameterization derived from a sub-set of the measurements was tested. This parameterization is given in Section 4.7. of this book. Hourly R_c values were parametrised and accordingly V_d values estimated using R_a and R_b estimates. It was found that with this parameterization, 44% of the variance in parametrised V_d versus the measured V_d was accounted for, with no systematic differences. An example of the comparison between parametrised and measured R_c is given in Figure 7.1. This parameterization is probably representative for Dutch environmental conditions. At Elspeetsche Veld annual average NH_3 concentration equals that of SO_2 (see also Section 3.5 about co-deposition). The parameterization might be improved with better information about surface wetness, its origin (dew, fog, rain, etc.) and its chemical composition, and about stomatal behaviour of heather plants.

Hicks *et al.* (1987) and Wesely (1989) presented parameterizations of R_c SO_2 for North American conditions. When these parameterizations are applied for the heathland dataset, too low V_d values are calculated in comparison to measured values. R_c values resulting from their parameterizations are much higher than those obtained from measurements and from Eqn (4.1). The main differences are found for periods with wet surfaces due to rainfall.

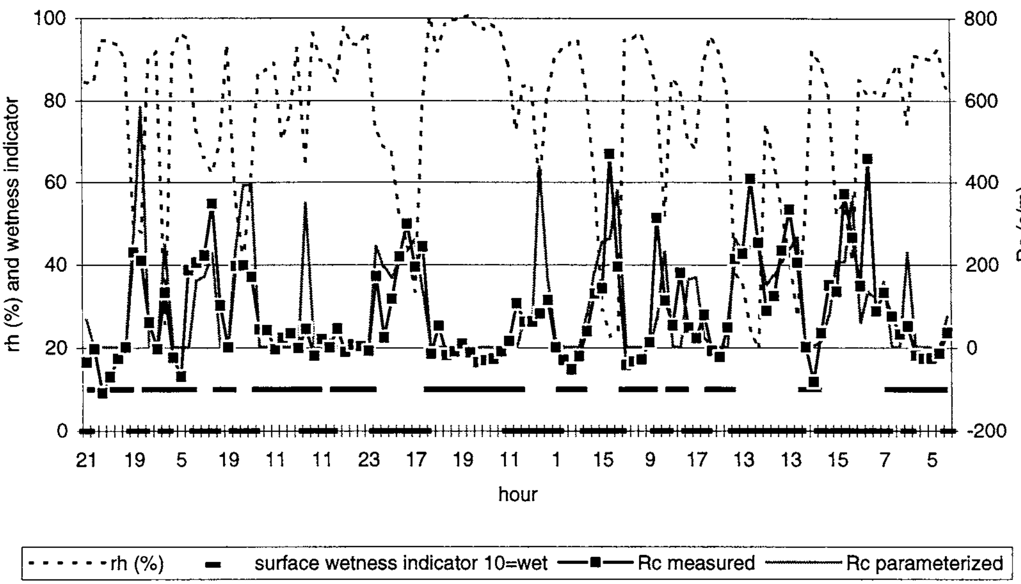


FIGURE 7.1 Time series of parametrised and measured R_c for SO_2 at Elspeetsche Veld.

Annual averages

There is a problem in estimating annual averages because by selection most data have been rejected (over 70%). Application of the rejection criteria results in overestimation of the annual average dry deposition velocity and dry deposition flux, since only periods with concentrations above the detection limit and with appreciable turbulence present were selected. However, it was found that data rejection does not leads to bias in R_c values. This was investigated by comparing the relative frequency distributions of the most relevant parameters used to derive surface conditions for the selected and the rejected dataset (Erisman *et al.*, 1990). R_c values are therefore expected to be similar for both the selected and total dataset.

The annual average dry deposition flux and V_d can be obtained from the resistance analogy by calculating R_a and R_b from the observations of u, T and R_n and taking the parameterization for R_c (Equation 4.1) corresponding the appropriate conditions. The annual average flux and deposition velocities for the complete datasets calculated using the parametrised R_c are listed in Table 7.4. In Table 7.4 uncertainties in deposition parameters are given. The estimate of the uncertainty in R_c is based on error propagation using uncertainties in all measured quantities to determine its value (see Section 3.1.1. and Erisman *et al.*, 1990; 1993; Erisman and Duyzer, 1991; Mennen *et al.*, 1992,. It is shown that the improved system (new monitors) yields more accurate deposition parameters than the system with 'old' SO_2 monitors (i.e. smaller uncertainties). Differences in R_a and R_b are the result of the different conditions during the years.

TABLE 7.4 Annual average dry deposition parameters for SO_2 at Elspeetsche Veld (4 m height). For the resistances the harmonic averages (± uncertainty estimates) are presented.

Parameter	'Old' monitors	'New' monitors	Dimension
z_0	4	4	cm
C	7.5	6.9	$\mu g\ m^{-3}$
R_a	43 ± 62	36 ± 45	$s\ m^{-1}$
R_b	23 ± 15	20 ± 28	$s\ m^{-1}$
R_c	47 ± 65	6 ± 8	$s\ m^{-1}$
V_d	0.8 ± 0.4	1.0 ± 0.1	$cm\ s^{-1}$
F	300 ± 270	250 ± 125	$mol\ ha^{-1}\ a^{-1}$

NO_2

The fully automated instrumentation to monitor the NO_2 flux was operated for a two month period (Duyzer, 1991). Some measurements during periods with easterly winds over a nearby road were rejected because of a possible interference due to NO_2 emissions from the traffic. Nevertheless, a large dataset consisting of some 1100 twenty-minute averages remains.

The deposition velocity obtained from these measurements ranged from 0.1 to 0.4 cm s^{-1}. A weak diurnal cycle was observed. The R_c values correspond to those estimated based on stomatal behaviour, and ranged from 100 to 200 s m^{-1} during the day to 700 s m^{-1} at night (see Figure 7.2). These results are consistent with those found in the literature (e.g. Duyzer *et al.*, 1990; Fowler *et al.*, 1991). The NO_2 flux for this two month period was estimated 500 mol ha^{-1} a^{-1}. No annual average flux can be obtained from these measurements because conditions cannot be regarded representative for a year.

NH_3

Thermodenuder measurements

From April 1989 to April 1990, NH_3 gradients at the heathland were measured by KEMA at two levels with an automatic analyser equipped with two thermodenuder instruments. A description of the apparatus and the measuring set-up is given in van den Beld and Römer (1990), Erisman *et al.* (1991) and Erisman (1992). In order to eliminate systematic differences in the concentration gradient, a quality programme was initiated (van den Beld and Römer, 1990; Erisman, 1992). During several days of the year, both thermodenuders measured at the same height (1 m). In this way, correction factors can be obtained for systematic differences between concentrations measured with the two thermodenuders. Furthermore, every two weeks the two thermodenuders were exchanged in order to measure at the other height. From the quality control tests, it was concluded that the error in the individual measurements was too large to use single gradients obtained with the two denuders for flux estimates (Erisman *et al.*, 1991; Erisman, 1992). Individual measurements could not be used for estimation of deposition parameters. However, deposition parameters were obtained by using yearly average diurnal variations of the concentration gradient and of meteorological data. Because of the regular exchange of the tubes between two heights, the corrections obtained from the quality control tests and a de-trending procedure, the errors were minimised when the gradients were averaged over the whole year (Erisman *et al.*, 1991; Erisman, 1992).

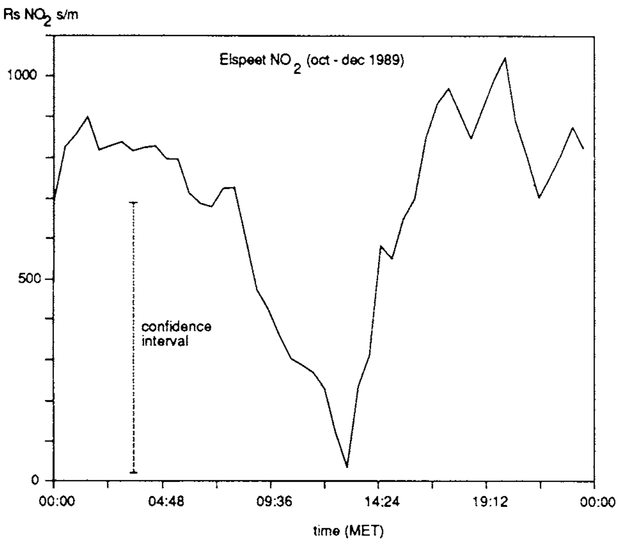


FIGURE 7.2 Averaged R_c for NO_2 over heathland (Elspeetsche Veld) in autumn 1989 (Duyzer *et al.*, 1991).

From the annual average diurnal variation of the concentrations and the annual average meteorological data the stability corrected concentration gradient c_* has been calculated based on the method described by Erisman (1992). From u_* and c_* the annual average diurnal variation of the flux and deposition velocity at a height of 4 m can be calculated for NH_3. By using annual averages instead of data for each measuring period an error is introduced because of correlation's between some parameters (between e.g. V_d and c). The magnitude of this error for NH_3 is unknown, because of lack of measurements to test the influence of these correlation's. For SO_2, this error was found to be small (Erisman *et al.*, 1990). The annual average diurnal variations of F and V_d are presented in Figure 7.3. A strong diurnal variation can be observed for both F and V_d. The annual average flux is 850 mol ha^{-1} a^{-1} and the annual average V_d is 0.8 cm s^{-1}. This is in agreement with similar measurements over heat land (Duyzer *et al.*, 1987; Sutton *et al.*, 1992). It must be emphasised that despite the minimisation of the errors, the uncertainty in these values is very large. Furthermore, separate averaging of meteorological data and concentration gradients over the year increases the uncertainty in the annual average F and V_d.

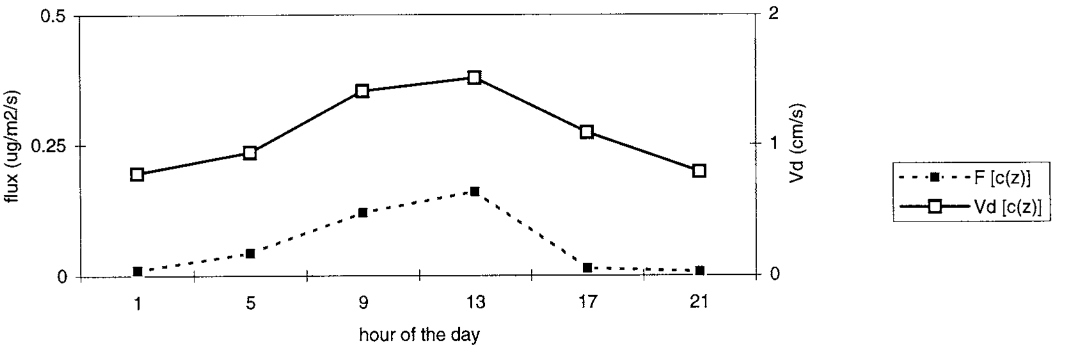


FIGURE 7.3 Annual average diurnal variations of F and V_d for NH_3.

Continuous flow denuder measurements

During three months at the end of 1991, continuous NH_3 gradients were monitored using continuous flow denuders (Wyers *et al.*, 1992; Erisman and Wyers, 1993). The measurements were attuned to the SO_2 gradients to yield average NH_3 concentration gradients for the same time intervals. The results of the simultaneous measurements of the two gases were used to derive indications for their possible interaction in the deposition process (see Section 3.5 and Erisman and Wyers, 1993). It was found that dry deposition of NH_3 is generally determined by turbulent transport with low R_c values of 15 s m^{-1} on average. The low R_c values are related to surface wetness which occurred most of the time. Surface wetness can be the result of either precipitation, fog or dew (visible or macroscopic wetness) or of high (>80%) relative humidity (microscopic surface wetness).

The changes in surface resistances of NH_3 match changes in relative humidity (see Figure 7.4). During very dry periods with rh<60%, high positive (deposition) or negative (emission) R_c values for NH_3 were observed. Upward fluxes of NH_3 were observed only during daytime. At higher relative humidities the surface resistances for NH_3 become small. An increase in rh from 30% to 95% was accompanied by a change from negative (emission) to near-zero values.

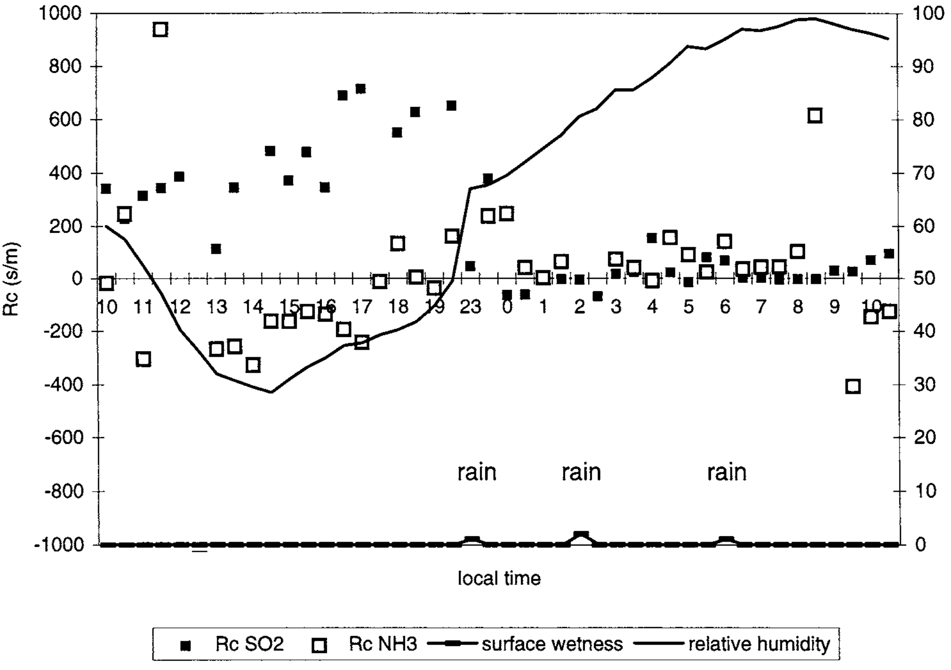


FIGURE 7.4 Surface resistance for SO_2 and NH_3, relative humidity and surface wetness indicator for the Elspeetsche Veld site, 14-15 September 1991 (Erisman and Wyers, 1993).

After rain or fog events volatilisation of NH_3 from water layers was sometimes observed. At temperatures below zero degrees centigrade at ground level, R_c values were observed to increase to several hundreds s m^{-1}.

Throughfall and stemflow

Bulk precipitation, throughfall and stemflow were sampled from 17 April 1989 to 17 April 1990 (Bobbink *et al.*, 1990; 1992). Rainfall was recorded until the end of 1989 with a pluviograph. Throughflow fluxes, defined as the sum of throughfall and stemflow, were found to be higher than bulk precipitation for all measuring periods. Throughflow fluxes of nitrate were relatively high in dry periods in May and in July. These periods corresponded with observed smog episodes of nitrogen dioxide in the summer of 1989 (RIVM, 1990). Throughflow fluxes of sulphate and ammonium were relatively high in autumn (October and November). This is probably related to wet Calluna canopies in this period leading to increased deposition (Bobbink *et al.*, 1992). In January and February 1991 high fluxes of sea-salt components were measured. Strong north-westerly winds, transporting marine air over the country, were observed in these months.

The annual bulk and throughflow fluxes of sulphate, ammonium and nitrate are shown in Table 7.5. The two-weekly fluxes of throughflow and bulk precipitation are given in Bobbink *et al.* (1990). Since foliar uptake or loss of sulphate was found to be negligible in heathland vegetation, the annual throughflow flux of SO_4^{2-} is considered to represent the total atmospheric deposition of sulphur to the vegetation. Furthermore, it was demonstrated that ammonium and nitrate in throughflow did not leach from the Calluna canopy. In contrast, foliar uptake of NH_4^+ by Calluna has been observed (Bobbink *et al.*, 1990; 1992).

TABLE 7.5 Annual average SO_4^{2-}, NO_3^- and NH_4^+ throughflow fluxes and bulk deposition (mol ha^{-1} a^{-1}). The standard deviation is given between brackets (Bobbink *et al.*, 1990).

	SO_4^{2-}	NH_4^+	NO_3^-
Bulk deposition	355 (9)	845 (42)	380 (3)
Throughflow	1035 (108)	2145 (311)	935 (145)
Interception deposition	680	1300	550

Comparison of atmospheric deposition estimates and throughfall measurements
The comparison is made for throughfall fluxes of SO_4^{2-} and NH_4^+ (Bobbink *et al.*, 1991). These fluxes comprise wet deposition and dry deposition of gases and aerosols. Only micrometeorological measurements of SO_2 and NH_3 fluxes were made. Aerosol data are lacking. Fluxes for SO_4^{2-} and NH_4^+ aerosol deposition were estimated by inference from concentrations obtained from the National Air Quality Monitoring Network and V_d estimates from u_* values (Wesely *et al.*, 1985; Erisman, 1992; 1993A). These aerosol measurements represent the fraction with small particle diameter (0.5 - 2.5 μm). The large particle fraction is not measured. However, this fraction is included in the throughfall. Thus throughfall fluxes have to be corrected for the contribution of this fraction according to Erisman (1992; 1993B). The throughfall data were also corrected for the dry deposition of gases and aerosols to the funnels of the devices (Erisman, 1993B).

The yearly average (corrected) net throughfall flux for SO_4^{2-} was 500 mol ha^{-1} a^{-1} and the dry deposition flux amounted to 350 mol ha^{-1} a^{-1}. The yearly average net throughfall flux for NH_4^+ corrected for foliar uptake (Bobbink *et al.*, 1992) was 1180 mol ha^{-1} a^{-1} and the dry deposition flux amounted to 930 mol ha^{-1} a^{-1}. The correlation between the two estimates on a monthly basis is significant (R=0.50 for SO_4^{2-} and R=0.52 for NH_4^+, p<0.05). The atmospheric deposition estimates tend to be somewhat lower. The agreement between net throughfall and dry deposition estimates is reasonable, provided that the corrections are made for the contribution of neutral salt deposition and of dry deposition on the sampling surfaces (Erisman, 1993B). Net-throughfall was somewhat higher for both SO_4^{2-} as well as for NH_4^+.

Co-deposition of SO_2 and NH_3

Simultaneous measurements of SO_2 and NH_3 concentration gradients were made (Erisman and Wyers, 1993). Furthermore, throughfall measurements of SO_4^{2-} and NH_4^+ were collected. These data might provide information on the process of co-deposition between SO_2 and NH_3.

Throughfall fluxes compared to atmospheric deposition

Bobbink *et al.* (1992) concluded from their throughfall measurements made at Elspeetsche Veld that stoichiometric co-deposition for SO_4^{2-} and NH_4^+ occurred. The conclusion was based on the observation that the slope of the regression equation of the throughflow fluxes of NH_4^+ and SO_4^{2-} is near to 2 with a regression coefficient of 0.91. In the net-throughfall fluxes this ratio is also found. Monthly mean dry deposition estimates of SO_2 and NH_3 were calculated using the resistance analogy, together with concentration measurements. Aerosol deposition was estimated as described in Section 7.3. The ratio of monthly mean dry SOx to dry NHx fluxes (aerosols included) obtained from the micrometeorological measurements does not differ from 2 (Erisman *et al.*, 1991; Erisman, 1992). However, the scatter is larger than that in throughfall.

The correlation that was found for the atmospheric fluxes of SOx and NHx is, in the present model formulation, not resulting from the co-deposition between SO_2 and NH_3 through pH coupling; neither the R_c parameterization of NH_3 nor that of SO_2 used for the calculation of these fluxes takes the pH coupling effect into account (see Chapter 4). The deposition velocities, however, are correlated because of the low R_c values used for both gases. V_d values are therefore determined by aerodynamic behaviour (R_a+R_b). The high correlation can thus be largely ascribed to the aerodynamic behaviour of the two gases. The calculated flux is the result of a concentration and a deposition velocity. The monthly average concentrations of NH_3 and SO_2 show a low correlation, although the annual average NH_3 concentration is twice that for SO_2. This might explain the 1:2 ratio found in throughfall.

The low R_c values for SO_2 and NH_3 are assumed to be caused by vegetation wetness. Furthermore, R_c values are different for day and night probably because of stomatal behaviour. An interdependence of R_c values for both gases will probably only have some influence on the fluxes under extreme conditions, i.e. very dry conditions, absence of NH_3 for R_c of SO_2, absence of SO_2 for the R_c for NH_3 or a large excess of NH_3 causing saturation of the surface and therefore increasing R_c for NH_3. The exact 2:1 relation found in throughfall samples might be the consequence of other processes such as evaporation of excess NH_4^+ in the samples.

Micrometeorological measurements of SO_2 and NH_3

So far there have only been a limited number of simultaneous micrometeorological measurements of NH_3 and SO_2, which are needed to test the importance of this process in the field. With the recent development of dry deposition monitoring systems for SO_2 and NH_3 (Erisman *et al.*, 1993; Mennen *et al.*, 1992; Wyers *et al.*, 1993), continuous measurement of the fluxes over extended periods and with sufficient accuracy has become possible.

The results of the simultaneous measurements performed in the autumn of 1991 clearly show the importance of surface wetness in the dry deposition process (Erisman and Wyers, 1993). However, the water layer composition affects the surface resistance of both gases. Under the environmental conditions prevailing in the Netherlands - commonly wet surface conditions - SO_2 and NH_3 are deposited with negligible surface resistances. A high flux of one gas relative to the other can increase its surface resistance through saturation of the surface and lower the resistance for the other gas. When NH_3 is present in excess over acid or acid-forming species, the surface resistance for NH_3 is mostly higher than zero, usually round 100 s m^{-1}, which is due to an increase in the pH of surface water layers. Analogous behaviour has been observed for SO_2 as a result of a decrease in the pH of surface water layers.

During dry periods (low *rh*), SO_2 and NH_3 surface resistances are determined by stomatal uptake. In very dry periods, small emission fluxes of NH_3 are observed. However, such conditions are not frequently observed in the Netherlands. At temperatures below zero, when the surface is frozen, R_c values of the two gases increase up to several hundreds s m^{-1}. Fog composition has a large influence on R_c values, resulting in both high and low values.

It is concluded that under the environmental conditions prevailing in the Netherlands, co-deposition is not an important process in the dry deposition of both gases. However, if the low R_c values found from independent measurements of SO_2 and NH_3 are the result of both gases being present in approximately the same concentrations during the measurements, then co-deposition is always important in the Netherlands. In those areas where there is a large excess of one gas over the other, co-deposition might certainly play an important role.

7.1.4 CONCLUSIONS

A three year experiment for the determination of deposition fluxes to heathland was conducted at the Elspeetsche Veld involving several research groups. From these measurements, annual average deposition parameters for SO_2 were estimated. Two extensive series of measurements resulted in estimates of V_d (4 m height) of 0.8 and 1.0 cm s^{-1} respectively. An R_c parameterization for SO_2 was derived which corresponds well with observed resistances. R_c values are usually low, related to surface wetness. A clear difference in R_c values between day and night and dry and wet conditions was observed. Changes in R_c values match changes in relative humidity in dry conditions.

From NO_2 concentration gradient measurements (TNO), deposition parameters were derived, yielding V_d values between 0.1 and 0.4 cm s^{-1}. R_c values generally followed stomatal behaviour.

NH_3 dry deposition was measured using different methods, which were evaluated. The annual average flux measured using thermodenuders (KEMA) was 850 mol ha^{-1} a^{-1} and the annual average V_d (4 m height) was 0.8 cm s^{-1}. It must be emphasised that despite minimisation of errors, the uncertainty in these values is large. During three months continuous NH_3 concentration gradients were monitored using continuous flow denuders (ECN). The measurements were made for the same time intervals as the SO_2 gradient measurements. It was found that dry deposition of NH_3 is generally determined by turbulent transport with low R_c values. These low R_c values were related to surface wetness which occurred most of the time. Surface wetness can be the result of either precipitation, fog or dew (macroscopic wetness) or of high (>80%) relative humidity (microscopic surface wetness). The changes in R_c of NH_3 match changes in relative humidity. During very dry periods with rh<60%, high positive (deposition) or negative (emission) R_c values were observed. Upward fluxes were only observed during daytime.

The influence of deposition of NH_3 to the deposition of SO_2 and vice verse was investigated. The deposition velocities of the two gases are correlated because V_d values are generally determined by turbulent transport, with low R_c values. The high correlation can thus be ascribed to the aerodynamic behaviour of the two gases. Also the annual average NH_3 concentrations were twice that for SO_2. This might explain the 1:2 ratio in throughfall studies. It could not be established whether the low R_c values for both gases are the result of surface wetness or a combination of surface wetness and the fact that concentrations of both gases were approximately equal.

Fluxes of SO_4^{2-} and NH_4^+ measured with the throughfall method were compared with fluxes derived using micrometeorological measurements. The yearly average (corrected) net throughfall flux for SO_4^{2-} was 500 mol ha^{-1} a^{-1} and the dry deposition flux (SO_2 and SO_4^{2-} particles) amounted 350 mol ha^{-1} a^{-1}. The yearly average net throughfall flux for NH_4 was 1180 mol ha^{-1} a^{-1} and the dry deposition flux (NH_3 and NH_4^+ particles) amounted 930 mol ha^{-1} a^{-1}. The correlation between the two on a monthly basis is significant (R=0.50 for SO_4^{2-} and R=0.52 for NH_4^+, n=12, p<0.05). The atmospheric deposition estimates tend to be somewhat lower.

7.2 THE UTRECHTSE HEUVELRUG EXPERIMENT ON THE IMPACT OF CANOPY STRUCTURE AND FOREST EDGE EFFECTS ON DEPOSITION

7.2.1 INTRODUCTION

Dry deposition depends heavily on the aerodynamic properties of the underlying surface. The supply of gases and particles from the free atmosphere to the receptor surface will be relatively high in case of 'rough' forest canopies exerting a large drag force on moving air masses. Especially tall forest canopies will exert a large drag force. Open and very dense canopies, caused by sheltering effects, will exert relatively small drag forces. Somewhere between these two extremes one might expect an optimum canopy density in the sense that maximum turbulent exchange occurs. Actual dry deposition amounts onto forest canopies depend on the efficiency of individual canopy elements to capture or absorb gases and particles. Small, needle-like structures are found to be more efficient in collecting particles and cloud droplets compared to larger leaf-like structures. Forest stands with small canopy density experience high in-canopy wind speeds which may enhance transport considerably.

Results from surface wash experiments (Van Dam *et al.*, 1987; Lindberg *et al.*, 1988; Draaijers *et al.*, 1992) and deposition modelling (Lovett and Reiners, 1986; Meyers *et al.*, 1989) suggest that dry deposition of various constituents increases linearly with increasing *LAI*. However, model results in particular indicate that this increment may level off or even be slightly reduced at higher *LAI* values. The *LAI* of forest canopies is found to influence transport from the free atmosphere to the receptor surface, as well as quasi-laminar layer transport and surface resistance.

In northern and western parts of Europe, extensive uniform forested areas are not common. In Scandinavia, for example, forest landscapes usually consist of a spatial mosaic of several subsystems: forest stands of different composition, height and canopy structure, logging areas, peat bog areas and lakes (Wiman, 1988). In the Netherlands and several other parts of Europe, forests are usually relatively small and surrounded by vast agricultural areas (Bleuten *et al.*, 1989). Consequently, many forest edges and other transition zones exist which are found to be important in affecting the exchange of momentum and mass between atmosphere and forest vegetation on both local (Wiman and Ågren, 1985; Li *et al.*, 1990) and regional scales (Klaassen, 1992).

Compared to forest interiors, atmospheric deposition in forest edges will be larger due to local advection and enhanced turbulent exchange. Wind entering a forest edge induces a pressure gradient of which the magnitude will be determined by the drag force introduced by the edge. This drag force will largely depend on the leaf area density of the forest edge. Relatively high in-canopy wind speeds significantly increase transport in forest edges, through which especially the deposition of super-micron particles and cloud and fog water droplets will be enhanced (Wiman and Ågren, 1985). Particle resuspension through bounce-off or blow-off processes may be relatively intense near the very edge of a forest at extremely high wind speeds and low stickiness of the collecting surface (Slinn, 1971; Wu *et al.*, 1992).

As explained in Chapter 4, for gases like NO_2 and SO_2 stomatal resistance is known to be one of the controlling factors for dry deposition. Enhanced solar radiation and daytime temperatures in forest edges (Ranney *et al.*, 1978) may reduce stomatal resistance and consequently promote dry deposition. At the same time, enhanced evapotranspiration (Miller, 1980) and the additional water consumption by dense understory vegetation usually present in forest edges, may reduce the availability of soil moisture through which stomata close and stomatal resistance increases. Surface wetness is also found to be an important feature with respect to dry deposition, especially for NH_3 and SO_2 (Adema *et al.*, 1986; Erisman and Wyers, 1993). It may be hypothesised that trees in forest edges are less frequently covered with a water film due to enhanced evaporation (Ranney *et al.*, 1978). However, interception of fog and cloud water is more efficient in forest edges than in forest interiors (Weathers *et al.*, 1992), which again could increase the occurrence of water films.

Existing models describing transport and deposition of pollutants in forest edges suggest a very large horizontal depletion of super-micron particles downwind of a forest edge (Wiman and Ågren, 1985; Bosveld and Beljaars, 1987; Van Pul *et al.*, 1991). The associated horizontal deposition patterns show distinct maxima around the forest wall. Submicron particles and gases are found subject to relatively small depletion and edge effects. This is in agreement with ambient air concentration measurements (Dasch, 1987; Wiman and Lannefors, 1985), and deposition measurements in forest edges using surface wash methods (Potts, 1978; Hasselrot and Grennfelt, 1987; Draaijers *et al.*, 1988; Beier and Gundersen, 1989). Little is known on the deposition enhancement in forest edges in relation to the forest structure and the exposition of the edge to prevailing wind directions.

Regional scale and receptor-oriented deposition models (see Chapter 5) treat deposition as an one-dimensional (vertical) transfer to homogeneous surfaces with infinite length. By determining turbulent exchange parameters at a height of 50 m above the receptor surface, it is believed that enhanced turbulent exchange induced by local roughness transitions is sufficiently taken into account in the deposition estimates (Erisman, 1992; 1993). Advection processes are not considered in these models. To date, there is no technique available to adequately compensate for 'edge effects' as the underlying processes are still scarcely studied and the land-use data necessary are usually not available.

This section presents results of an extensive field study on the effects of canopy structure and edge aspects on deposition gradients in forest edges. On the basis of these results and information gathered on forest fragmentation in the Netherlands, the deposition to Dutch forests is estimated taking edge effects into account.

7.2.2 METHODS

Study area

To study the relationship between atmospheric dry deposition and canopy structure, an extensive throughfall monitoring programme was performed in the middle of 30 different forest stands (1-30, Figure 7.5). Forest edge effects were assumed negligible as the distance to the nearest stand edge always exceeded 50 m (five edge heights). Nine measurement sites were situated in Douglas fir (*Pseudotsuga menziesii* Mirb. Franco) stands, 10 sites in Scots pine (*Pinus sylvestris* L.) and 11 sites in oak (*Quercus robur* L.) stands. To study the spatial deposition variability within forest stands, very detailed throughfall monitoring was performed in one Douglas fir, one Scots pine and one oak stand. Bulk precipitation was measured in five clearings scattered over the study area.

To study edge effects, throughfall was monitored at eight forest edges (A-H, Figure 7.6). Forest edge measurements were conducted in three European larch (*Larix decidua* Mill) stands, three Scots pine (*Pinus sylvestris* L.), one Corsican pine (*Pinus nigra* var. maritima) and one Norway spruce (*Picea abies* L.) stand. All edges were exposed to the prevailing southwesterly winds. More-or-less undisturbed fetch over heather, grassland or arable land was found in front of the edges. Bulk precipitation was measured in the vicinity of each forest edge.

All forest stands and forest edges investigated were located in a forested area in the central part of the Netherlands, called the 'Utrechtse Heuvelrug'. The Utrechtse Heuvelrug is an approximately 50-m high ice-pushed morainic ridge with dry, sandy and nutrient-poor podzolic soils. Large source areas of SO_2 and NO_x are located 200 km to the southeast (industrial Ruhr area) and 100 km to the southwest (Rotterdam port) of the Utrechtse Heuvelrug. NO_x also originates from local traffic on small roads crossing the Utrechtse Heuvelrug. The forested area is enclosed by two relatively large agricultural areas, which are called the 'Gelderse Vallei' and the 'Kromme Rijn area'. These areas are large sources of NH_3 due to ammonia volatilisation from animal manure. Within the forested area, some scattered farms with intensive animal husbandry also act as a source of ammonia.

It was expected that the 30 measurement sites used to study the relationship between atmospheric dry deposition and canopy structure were expected to be exposed to more-or-less similar air concentrations as they were situated within a radius of 1.4 km of each other. However, it was recognised that some local NH_3 air concentration gradients may have existed due to the location of the study area near two large ammonia emission areas and the situation of some intensive livestock farms within the study area. Moreover, small NOx air concentration gradients were expected near few thoroughfares crossing the study area, along with local air concentration gradients of Ca^{2+} and other base cation-containing particles near arable land and small unmetalled forest roads.

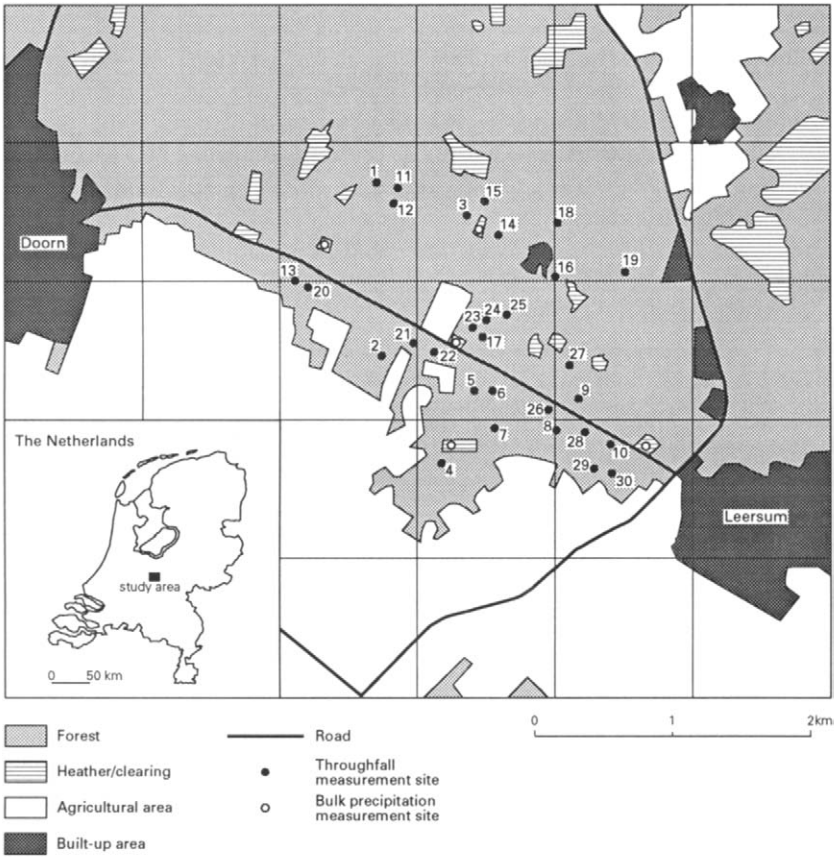


FIGURE 7.5 Location of the study area with the 30 throughfall measurement sites and five bulk precipitation measurement sites used to study the impact of canopy structure on dry deposition amounts.

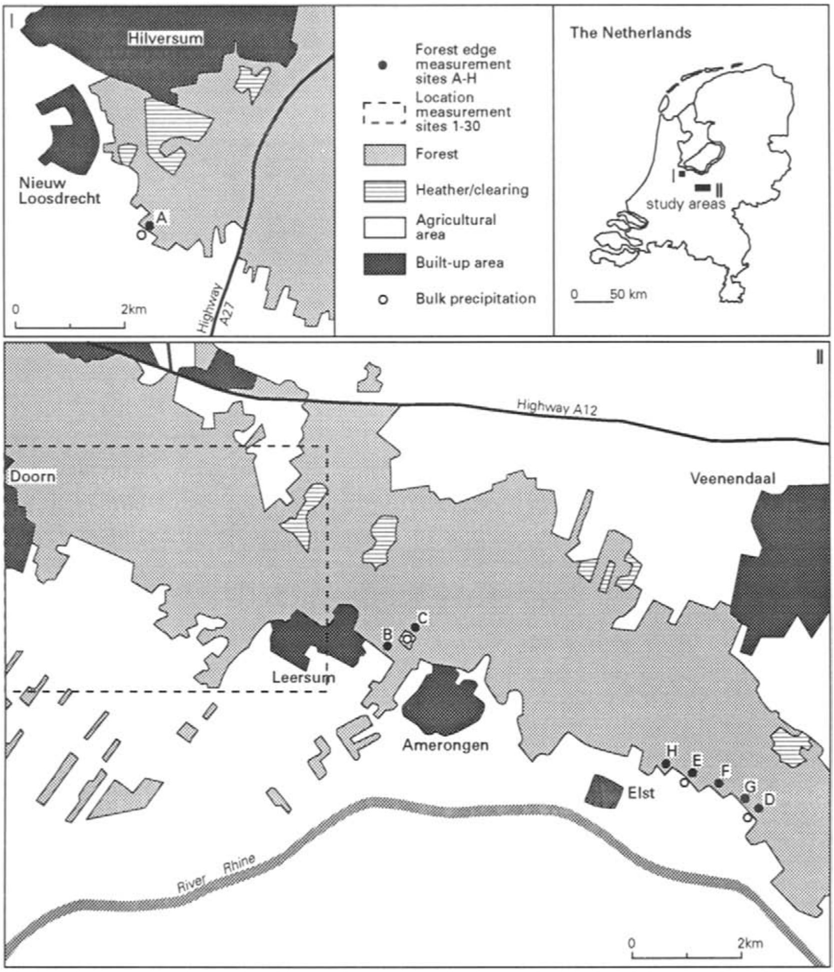


FIGURE 7.6 Location of the study area with the eight forest edge measurement sites and four bulk precipitation measurement sites used to study the impact of edge effects on dry deposition amounts.

Throughfall and bulk precipitation sampling procedure

Field procedures

In each of the 30 forest stands two throughfall gutters were placed near the stand centre. Each throughfall gutter was 5 m long and had a total collecting area of 0.054 m^2. They were placed at a slight angle of approximately 15° to the horizontal. Throughfall gutters were preferred to funnels because they integrate over a larger canopy area, yielding more representative estimates of the throughfall flux (Duijsings *et al.*, 1986; Beier and Rasmussen 1989; Ivens, 1990). A large number of funnels would have been needed to obtain similar results (Kimmins, 1973; Draaijers *et al.*, 1995). The spatial variability of throughfall fluxes in the middle of three different forest stands was studied by means of eight throughfall gutters. In the eight forest edges, throughfall gutters were placed at a distance of 5, 10, 20, 30, 40, 60, 80 and 150 m from each edge (up to 6.0-11.4 edge heights from the edge). Gutters at 150 m from the edge were only placed when the depth of the forest stand exceeded 200 m to be sure other roughness transitions had negligible impact on throughfall fluxes. Bulk precipitation was measured by two funnels positioned at a distance of at least three times the height of the nearest obstacle. Nearby obstacles will most probably still have had a certain impact on the air flow around the funnels and, consequently, on their collecting efficiency. The funnels were placed approximately 1.5 m above the ground surface to prevent soil splash. Each funnel had a collecting surface area of 0.017 m^2.

To avoid contamination, e.g. by insects and coarse litter fragments, throughfall and bulk precipitation were filtered (mesh width of filter = 250 μm) before entering a 10-*l* and 2-*l* reservoir, respectively. Only opaque reservoirs were used to avoid light penetration, which could promote algae growth. The materials used for sampling (principally polyethylene) were all chemically inert as far as ions were concerned. Gutters, funnels, reservoirs and filters were cleaned with de-ionised water after each sampling period. Throughfall and bulk precipitation samples were stored in the field no longer than one week to prevent biochemical transformations (Slanina *et al.*, 1990).

Throughfall and bulk precipitation at the 30 measurement sites and eight forest edges were measured continuously for one year. For the 30 measurement sites collection started on 17 May 1990 and ended on 30 April 1991. During that time period 18 throughfall and matching bulk precipitation collections were made in total. At locations B, C and D, collection started on 27 September 1989 and ended on 27 September 1990; 23 collections were performed, while at the locations E, F, G, and H, collection starting on 15 October 1990 and ended on 1 October 1991, with 16 collections in total. At location A, throughfall and bulk precipitation was measured continuously for two years (from 27-09-89 to 01-10-91; 39 collections). To achieve complete removal of dry deposited material from the canopy, water was collected, as far as possible, after showers bringing large amounts of rain (preferably >10 mm). However, the fact that such rain amounts are not common in the Netherlands and the condition that samplers were not allowed to stay in the field longer than one week regularly prevented this. The matching deposition periods were assumed to start immediately after the end of the last rain event of the previous collection. This information was gathered from results of a pluviograph situated near the measurement sites.

Laboratory procedures and data management

After collection, samples were stored in darkness at 5 °C. Samples were analysed for acidity (pH) and electric conductivity (EC) within 24 hours of sampling. Ion concentrations were measured within one week of sampling. Samples were analysed by colorimetry using a Skalar autoanalyser for ammonium (NH_4^+), nitrate (NO_3^-), sulphate (SO_4^{2-}), chloride (Cl^-), magnesium (Mg^{2+}), calcium (Ca^{2+}), orthophosphate (PO_4^{3-}) and bicarbonate (HCO_3^-). Furthermore, ion concentrations of potassium (K^+) and sodium (Na^+) were determined by flame photometry. In the period 7 January 1991 to 1 October 1991, Kjeldahl-N analyses were also performed on throughfall samples from the forest edges A, E, F, G, and H, collected at 5, 30 and 80 m distance from each edge, respectively. In this way, information was gathered on the organic nitrogen content in throughfall, as organic N was assumed equal to Kjeldahl-N minus N-NO_3^- and N-NH_4^+.

Directly after the laboratory analysis, measured ion concentrations were entered into the data management programme, DAVER (Klein Tank, 1990), in order to calculate ion balances. Through this approach, ion concentrations were directly verified and repeated within 24 hours, if necessary. Analytical errors were found to be small (< 5%). However, concentration measurements were less reliable when concentrations were below the detection limit of the analysing equipment; this sometimes occurred for SO_4^{2-}, PO_4^{3-}, HCO_3^-, Mg^{2+} and Ca^{2+} in bulk precipitation (analytical error ± 20%). All chemical analyses were performed at the Laboratory of Physical Geography at Utrecht University. The laboratory regularly participates in interlaboratory tests to ensure good quality concentration measurements.

Interpolation of missing values

Approximately 10% of all throughfall and bulk precipitation data (concentrations and/or amounts) were missing because of damaged collecting equipment due to vandalism (humans and/or rabbits) or gales, or was not usable due to bird droppings onto the collectors. Bird droppings were detected in the field or could be ascertained by unusually high pH, PO_4^{3-}, K^+ and/or HCO_3^- concentrations in the samples. These missing values were interpolated using the remaining data set through the very strong spatial correlations observed between two throughfall gutters or two bulk precipitation funnels situated close to each other. Due to the large size of the dataset, an interpolation method using ratios was preferred to the more time-consuming method using linear regressions between two gutters or funnels. Moreover, a comparison between the ratio method and the regression method using a small subset of the data did not yield significantly different results.

Hail and snowfall occurred from 20 January until 15 February 1991, and no single reliable throughfall and bulk precipitation collection could be made. For this reason, the method of spatial interpolation could not be applied. Precipitation volumes during this period were measured using slightly heated pluviographs. Bulk precipitation concentrations were interpolated using the fairly strong (temporal) correlation observed between precipitation concentrations and volume. Throughfall volumes were estimated from the strong (temporal) correlation found between throughfall and bulk precipitation volumes, whereas throughfall concentrations were assumed equal to the mean ratio between throughfall and bulk precipitation concentrations in the winter period. The latter interpolation procedure probably yields an underestimate of the throughfall concentrations as, compared to winter averages, relatively high air concentrations were observed in the dry period preceding the snowfall (RIVM, 1991). All round, this interpolation procedure does not seem very reliable, but there was no better alternative available; the error introduced in the annual mean bulk precipitation and throughfall fluxes was assumed to be smaller than 5%.

Calculation of deposition fluxes

After interpolation procedures, bulk precipitation fluxes for each sampling period were calculated by multiplying rainfall concentrations with rainfall amounts. Similarly, throughfall fluxes were calculated by multiplying throughfall concentrations with throughfall volumes. To obtain wet deposition amounts, bulk precipitation fluxes were corrected for dry deposition onto the collectors using the correction factors of Ridder *et al.* (1984) (see Table 3.1). The dry deposition amounts were subsequently estimated by subtracting wet deposition amounts from throughfall fluxes (= net throughfall). Throughfall fluxes were not corrected for dry deposition directly onto the gutters because the amount of dry deposition is expected to be relatively small. By using net throughfall as an indicator for dry deposition, the contribution of cloud and fog water deposition and canopy exchange to net throughfall was neglected. Annual mean wet and dry deposition fluxes were computed by aggregating the fluxes calculated for single periods. Total deposition was calculated by summing wet and dry deposition fluxes.

Canopy and edge structure measurements

Around each throughfall gutter, an inventory of relevant canopy structure characteristics was made in order to determine the aerodynamic roughness of the canopy, the collecting efficiency of individual canopy elements and the total collecting surface area. From each tree of which the crown was (partially) located within a 100 m^2 plot around the gutter (Figure 7.7), the following information was gathered through field measurements or model calculations using the canopy structure model TREE especially designed for this purpose (Van der Zanden and Sluyter, 1990):

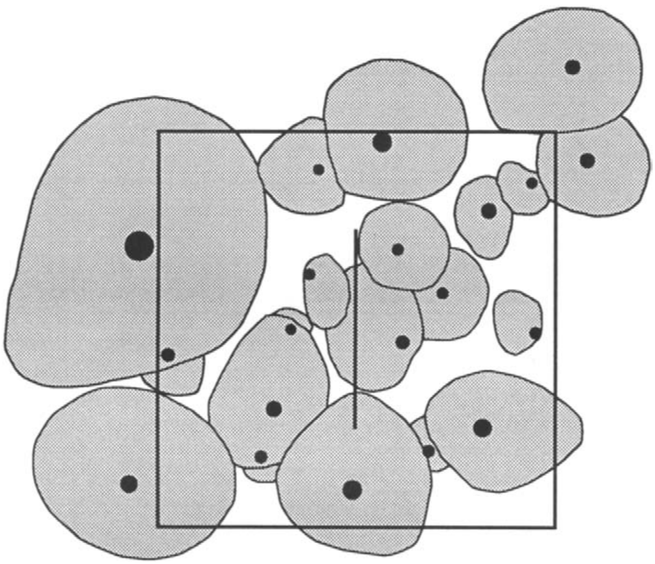


FIGURE 7.7 An example of a measurement site showing the area (10 x 10 m) around the throughfall gutter in which tree and stand structure characteristics were determined. The throughfall gutter, indicated as a black bar, is located in the centre of the plot. Grey areas mark the vertical crown projection of individual trees, whereas stem bases are indicated as black circles. The thick line indicates the boundary of the 100 m^2 plot.

Location of stems, stem density and stem diameter
The exact location of the stem base within (or outside) the plot was measured with help of a measuring tape (measurement error ± 10 cm). The number of trees within the 100 m^2 plot was computed as the sum of stems weighted for the amount of the crown volume of each tree located within the plot. The stem diameter at breast height was measured in two directions perpendicular to each other using a large marking gauge (measurement error ± 0.5 cm).

Crown top, crown base and periphery height, and crown radius
The height of the crown top, living crown base and periphery height (where the crown has its maximum horizontal radius) was measured in the field using a standard tree-height measuring device based on goniometry (measurement error ± 5 %). The crown depth was defined as the crown top minus the crown base. The radius of the crown at periphery height was measured in eight wind directions with help of a compass, a long pole and a measuring tape (measurement error ± 10-20 cm).

Crown volume and crown silhouette area

To compute the crown volume and silhouette area (= frontal crown area) of each individual tree, field information on the height of the crown top, crown base and periphery was used together with field information on the eight crown radii, and so-called 'crown shape exponents' describing the concavity or convexity of the crown above and below the periphery height, respectively (Koop, 1989). These shape exponents were selected in such a way that the resulting crown model best reflected the shape of the crown as observed in the field.

Leaf/needle area and leaf/needle area density

Several workers have reported a functional relationship between the total needle area of a tree and the water-conductive capacity of the stem (e.g. Waring *et al.*, 1982). To express this water-conductive capacity, usually sapwood area is used, although some workers have pointed out that also wood density, hydraulic conductivity and the number of annual rings must be taken into account (e.g. Espinosa Bancalari *et al.*, 1987). The slope of the relationship between needle area and sapwood area is found to differ with growing conditions (Whitehead, 1978) and tree species (Waring *et al.*, 1982). For the measurement sites in this study, growing conditions were similar since all trees were growing on poor, sandy podzolic soils with deep groundwater table and were exposed to equal climatic conditions.

A small tree borer was used to determine the sapwood area of Douglas fir, Scots pine, Corsican pine and Norway spruce trees. Two opposite wood cores in the direction of the thickest stem diameter were taken into account for possible asymmetrical positioning of the sapwood. For Scots pine, Corsican pine and Norway spruce trees, sapwood appeared as glassy tissue if the core was held towards the light. For Douglas fir trees, a clear colour distinction could be made between heartwood and sapwood. Except for sapwood, the number of annual rings was also counted to compute tree age. The sapwood area was calculated according to π^* (total stem radius from centre to phloem)^2 minus π^* (mean radius covering heartwood)^2.

In January 1991 needles were destructively sampled from three Scots pine, three Corsican pine, three Norway spruce and three Douglas fir trees to determine the relationship between sapwood and needle areas. Special care was taken to ensure that the trees destructively sampled covered the range of tree heights present at the measurement sites. Due to practical reasons only 20% of all needles were sampled and weighed after drying at 70°C for 48 h. Weight of the remaining 80% was extrapolated on the basis of a count of branches left intact on the trees sampled. To compute specific needle area (i.e. surface area of fresh needle divided by its corresponding dry weight), a subsample of fresh foliage was used, as foliage was found to shrink as much as 25% in drying (Waring *et al.*, 1982). Total needle area was calculated by multiplying dry weight of all needles by specific needle area. The needle area density of an individual tree was computed by dividing total needle area by crown volume.

Although only three trees were destructively analysed from each species, clear and significant relationships were found between total needle area (*TNA*, in m^2) and sapwood area (*SA*, in cm^2):

Scots pine:	[*TNA*] = 18.85 + 0.230 * [*SA*]	(*R*=0.9996; *R*= 0.018)
Corsican pine:	[*TNA*] = -3.20 + 0.388 * [*SA*]	(*R*=0.9979; *R*= 0.041)
Norway spruce:	[*TNA*] = -8.16 + 0.895 * [*SA*]	(*R*=0.9989; *R*= 0.030)
Douglas fir:	[*TNA*] = 2.67 + 0.603 * [*SA*]	(*R*=0.9970; *R*= 0.049)

For deciduous trees, the relationship between surface area and sapwood area is found to be less exact (Rogers and Hinckley, 1979). Therefore, the leaf area of oak and European larch was estimated by litterfall collections. Litterfall collections were made using three litter traps per 100-m^2 plot. Each littertrap had a collecting surface area of 1 m^2 arranged in a row in the centre of a plot beneath the throughfall gutter. Litterfall collections were made during the autumn until no more leaves remained on the tree branches. Total leaf area collected with the litter traps was calculated by multiplying specific leaf area by the total dry weight. This value was multiplied by 100/3 to compute total leaf area per 100-m^2 plot. Thus, a mean plot leaf area was obtained but no information was gathered on leaf areas of individual trees. Unfortunately, this method could not be applied near forest edges. At forest edges, leaf collection will not be representative for the canopy leaf area right above the gutter due to relatively high horizontal wind speeds within and under the canopy. For this reason, the leaf litter collection method for forest edges was only applied at 150 m from the edge.

Internal crown cover, crown area and crown projection area

The internal crown cover of individual trees was visually estimated and expressed in percentage classes of 5%. Although this technique is rather subjective, repeated estimates showed fairly consistent results, even when the observations were made by different observers. Most estimates differed only by 10% percentage class. For oak and European larch trees, the internal crown cover was estimated for both foliated and unfoliated conditions. The crown area of each individual tree was computed using the eight crown radii measured in the field. This was done by interconnecting the ends of the eight crown radii by a straight line, thereby forming a polygon with eight corners. The area of this polygon was assumed to equal the crown area. The crown projection area was calculated by multiplying calculated crown area by the internal crown cover measured in the field.

Crown projection area estimated from scanned images

Because the visual estimation of the internal crown cover was rather subjective, panchromatic images by means of photography were also used to determine crown projection areas. For the oak and European larch stands, both winter and summer images were taken to account for the seasonal variability of the crown cover. Images were only made on days when the sky had a uniform cloud cover. These weather conditions made the canopy appear as black against a white background. Days with clear skies were not useful because direct sunlight illuminated canopy foliage, causing an underestimation of the total canopy projection area. An image of the canopy right above the middle of each throughfall gutter was made using a 28-mm standard lens pointing vertically, making sure the zenith angle of the camera equalled 0°. Such a wide-angle lens has longitudinal and latitudinal recording angles of 37.5° and 25°, respectively. Images were scanned using ERDAS digital processing techniques. The number of black pixels divided by the total number of pixels was used as an estimate of the crown projected area. Three rectangular areas of different size were scanned from each image, namely the full-size image, a medium-size image (corresponding to longitudinal and latitudinal recording angles of 27.5° and 15°, respectively) and a small-size image (corresponding to recording angles of 22.5° and 10°, respectively). Thus, no information was gathered on crown projection areas of individual trees. The actual canopy area scanned differed for each image. Except for recording angles, the actual canopy area scanned depends on the mean tree height. The actual canopy area scanned can be computed as [2 h $\tan(\alpha)$] by [2 h $\tan(\beta)$], in which α and ß represent recording angles and h the mean tree height.

Computations for the whole 100-m^2 plot

Besides the mean stem diameter and mean height of the crown top, crown base and periphery for each 100 m^2 plot, the total crown volume, total silhouette area, total crown projection area and total leaf/needle area of each 100 m^2 plot were also computed with the restriction that overlapping crown volumes, silhouette areas and projection areas were only counted once. Moreover, crown volumes, silhouette areas, crown projection areas and leaf/needle areas located outside the 100 m^2 plot were excluded.

Estimation of the aerodynamic roughness of the canopy

The roughness length is often used to characterise the aerodynamic roughness of a forest canopy (Thom, 1971). Due to the large number of measurement sites, it was impossible to estimate the roughness length in practice from measurements on the logarithmic wind profile. For this reason, the roughness length of the forest canopy above each throughfall gutter was estimated using the geometrical model of Lettau (1969). For Douglas fir, Scots pine, Corsican pine and Norway spruce trees, a drag coefficient of 0.35 was assumed (Leonard and Federer, 1973). For foliated conditions in the summer period, the drag coefficient of oak and European larch trees was also assumed to be equal to 0.35, whereas for unfoliated conditions in the winter period, a drag coefficient of 0.13 was assumed (Kruijt, 1986). The effective height in Lettau's model is calculated by h-d with h equal to the mean tree height and d the zero plane displacement. The zero plane displacement varied between 0.57 h and 0.87 h, depending on the total crown projection area (cpa) within the 100 m^2 plot:

$$d/h = 0.57 + 0.3 * (cpa/100) \qquad [7.1]$$

To compute the roughness length of a canopy according to the geometrical model of Lettau, information on silhouette areas and number of trees within the 100 m^2 plot was also used.

Estimation of the collecting efficiency of individual canopy elements
Parameters expected to influence the collecting efficiency of individual leaves/needles include mean projected surface area and diameter (width). For this reason, 150 Douglas fir and 150 Scots pine needles were sampled and measured as accurately as possible using a small marking gauge. As the shape of Douglas fir needles was found to be practically flat, only the length and the maximum diameter were recorded. The shape of Scots pine needles was regarded as a half-cylinder lengthwise of which length, minimum diameter, and maximum diameter were recorded. The dimensions of oak leaves were not measured because the difference in collecting efficiency with Douglas fir and Scots pine needles was obvious.

Other parameters related to the collecting efficiency, such as 'stickiness' or 'hairiness' of the leaf/needle surface, were also recorded. However, the clustering of leaves/needles was not quantified although it was recognised that this may be an important parameter with respect to the collecting efficiency of individual leaves/needles.

Estimation of the collecting surface area of the canopy
The most direct measure for the total collecting surface area of a canopy is the *SAI*. Because leaves/needles usually contribute more than 85% to the total surface area of trees (Halldin, 1985), the LAI was used in this study. A lower percentage will probably hold for trees with leaf/needle loss as a result of reduced vitality. The total crown projection area may be regarded as an indirect measure for the collecting surface area. Besides the field estimates, estimates from the scanned photographs were also used. Total crown volume and mean crown depth also reflect to some extent the collecting surface area of a canopy.

Estimation of edge structure characteristics
Parameters important for the aerodynamic properties of forest edges include relative height, edge porosity, and fetch and roughness of the upwind terrain. The relative height of the eight forest edges was computed as the mean tree height of the forest minus the height of the vegetation in front of the edge. Special attention was paid to irregular tree height with distance to the forest edge because this was also to some extent supposed to influence edge aerodynamics.

The porosity of a forest edge (i.e. the proportion of open area (background sky) to the total edge area) was estimated by scanning panchromatic images made at both one- and two-edge heights into the forest interior, looking outside. Forest edge images were processed in a similar manner as described for canopy images. Edge porosity was also estimated visually, in a way comparable to the estimation of the internal crown cover of trees. Edge porosity was determined 2 m above the forest floor, in this way it was more representative for the lower levels of the edge (trunk area). This method for characterising edge porosity turns the forest edge in a two-dimensional plane, through which objects in the foreground look larger than those in the background. Moreover, only the two-dimensional gaps, and not all the spaces through which the wind flows across the forest edge, are determined. Therefore the optical porosity estimated is not similar to the real porosity of the forest edge (Nord, 1991). Edge porosity was also characterised by the stand leaf/needle area density defined as the total leaf/needle area divided by the total stand volume (i.e. plot area times mean tree height). The drag force a forest canopy exerts on moving air masses may also be characterised by the stand silhouette area density computed as the total silhouette area divided by the total stand volume. Crown leaf/needle area densities and crown silhouette area densities were similarly computed. For this, total canopy volumes (i.e. plot area times mean crown depth) were used instead of total stand volumes.

Recording of the land use in front of the forest edge allowed a rough estimate of the fetch and the roughness length of the upwind terrain to be made. A terrain classification made by Wieringa (1992) was adopted to obtain roughness lengths. In this classification, the roughness length of fallow ground is averaged to 0.0025 m, that of short grass to 0.019 m, that of long grass and heather to 0.04 m, that of low mature agricultural crops to 0.065 m, and that of high mature agricultural crops to 0.15 m.

7.2.3 THE IMPACT OF CANOPY STRUCTURE: RESULTS AND DISCUSSION

Evaluation of canopy structure parametrisation

Results of the canopy structure measurements are summarised for the 30 measurement sites in Table 7.6. A discrimination has been made between parameters related to the aerodynamic roughness, and parameters reflecting the collecting surface area of the canopy. The classification of a single parameter into a particular category was sometimes arbitrary. The roughness length as estimated with the geometrical model of Lettau (1969) was, on average, 2.1 m and ranged from 1.1 m to 4.0 m. Input parameters for the geometrical model of Lettau revealed large spatial variability: mean tree height was between 12.4 m and 24.1 m, stem density between 340 and 1300 trees per ha and silhouette area between 7100 and 20,900 m^2 ha^{-1}. Uncertainties in the estimate of the roughness length were primarily associated with the rather badly calibrated drag coefficient.

The leaf area index was, on average, 6.7 and ranged from 2.4 to 12.0. Other parameters related to the collecting surface area of a canopy also showed wide ranges: crown projection (field estimates): 17.7%-71.8%, crown projection (estimated from scanned 'full-sized' images): 48.1%-86.5%, crown volume: 13,700-85,200 m^3 ha^{-1} and crown depth: 3.9-18.8 m. Crown projection estimates from scanned 'full-sized' images were similar to crown projections estimates from scanned medium-sized' and 'small-sized' images. A strong relationship was observed between crown projection estimated from scanned images and field estimates (R=0.79; $p<0.001$; n=30) but estimates from scanned images were significantly larger than field estimates. This was most probably the result of including the stem areas in the estimates of scanned images, whereas in the field estimates stem areas were neglected. A strong relationship was found between the difference of both estimates and stem density (R=0.67;

$p<0.001$; $n=30$). Because the 30 measurement sites had rather low vitality indices (section on Canopy...measurements), trunks, branches and twigs may have contributed significantly to the total collecting surface area of the canopy. This introduces an uncertainty when using *LAI* as a measure for the collecting surface area of the canopy.

TABLE 7.6 Overview of stand structure characteristics of the 30 forest stands

	x	*sd*	*cv*	max	min	max/min
Aerodynamic roughness						
z_0 (m)[a]	2.1	0.6	0.30	4.0	1.1	3.62
Tree height (m)	18.0	2.6	0.14	24.1	12.4	1.95
Silhouette area ($m^2\ ha^{-1}$)	13040	3538	0.27	20896	7096	2.94
Stem density (ha^{-1})	686	243	0.35	1302	341	3.82
Collecting surface area						
LAI (-)	6.7	2.8	0.41	12.0	2.4	5.00
Crown projection[b], field (%)	40.9	12.9	0.31	71.8	17.7	4.05
Crown projection[b], image (%)	64.5	9.7	0.15	86.5	48.1	1.80
Crown volume ($m^3\ ha^{-1}$)	35510	16383	0.46	85160	13740	6.20
Crown depth (m)	9.0	4.4	0.49	18.8	3.9	4.82

x = mean, *sd* = standard deviation, *cv* = coefficient of variation

[a] For z_o, *LAI* and crown projection of oak, annual mean values are presented which were computed by averaging summer and winter values.

[b] For crown projection, field estimates and estimates from scanned 'full sized' images are presented.

The roughness length, leaf area and crown projection of oak stands differed greatly with the seasons (Table 7.6). This contradicted observations made by Dolman (1986), who reported negligible impact of leaf shedding on the roughness length of an oak stand. The drag coefficient used to compute the roughness length of the oak stands in the summer period was based on experiments done by Kruijt (1986), and equalled the drag coefficient used for Douglas fir and Scots pine trees (0.35). However, it may be hypothesised that the drag coefficient for oak trees will be smaller than that of coniferous trees because per unit surface area individual leaves initiate smaller drag forces compared to needles (Thom, 1968; Monteith, 1973). Assuming that a correct drag coefficient was used for the computation of the roughness length in the winter period, a summer drag coefficient of 0.20 should have been used to obtain summer roughness length values comparable to those in the winter period. However, the drag coefficient used for the winter period (0.13) was also subject to considerable uncertainty. The leaf area of Douglas fir and Scots pine stands will also show some variability with time but this variability was not quantified. Measurements at Speuld have indicated that the leaf area of Douglas fir stands may deviate up to 30% from the annual mean (Steingröver, 1993, personal communication).

TABLE 7.7 Overview of stand structure characteristics of single tree species

	x	sd	cv	max	min	max/min
Douglas fir (n=9)						
z_o (m)[a]	2.7	0.8	0.28	4.0	1.8	2.22
Tree height (m)	19.5	2.6	0.13	24.1	14.8	1.63
Silhouette area (m^2 ha^{-1})	14941	4011	0.27	20896	8755	2.39
Stem density (ha^{-1})	662	271	0.41	1302	433	3.01
LAI (-)	9.4	1.6	0.17	12.0	7.5	1.60
Crown proj. field (%)	54.1	11.0	0.20	71.8	39.9	1.80
Crown proj. image (%)	76.0	8.8	0.12	86.5	59.2	1.46
Crown volume (m^3 ha^{-1})	49491	18293	0.37	85160	22950	3.71
Crown depth (m)	8.3	1.8	0.22	10.5	5.9	1.78
Scots Pine (n=10)						
z_o (m)[a]	1.9	0.4	0.18	2.4	1.3	1.85
Tree height (m)	17.1	1.1	0.06	18.3	15.1	1.22
Silhouette area (m^2 ha^{-1})	9545	1442	0.15	11205	7096	1.58
Stem density (ha^{-1})	717	265	0.37	1112	407	2.73
LAI (-)	7.9	1.2	0.15	9.9	6.3	1.57
Crown proj. field (%)	31.2	9.0	0.29	44.9	17.7	2.53
Crown proj. image (%)	59.9	3.7	0.06	65.0	50.9	1.28
Crown volume (m^3 ha^{-1})	23584	8771	0.37	45200	13740	3.29
Crown depth (m)	4.9	0.7	0.14	6.0	3.9	1.54
Pedunculate oak (n=11)						
z_o, summer (m)[a]	2.2	0.5	0.23	3.2	1.4	2.23
z_o, winter (m)[a]	1.3	0.3	0.24	1.8	0.8	2.32
z_o, annual (m)	1.8	0.4	0.23	2.5	1.1	2.25
Tree height (m)	17.6	3.1	0.18	23.3	12.4	1.88
Silhouette area ($m^2.ha^{-1}$)	14663	1786	0.12	17239	11598	1.49
Stem density (ha^{-1})	677	218	0.32	1031	341	3.02
LAI, summer (-)	7.1	1.0	0.15	8.0	4.7	1.70
LAI, winter (-)	0.0	0.0	--	0.0	0.0	--
LAI, annual (-)	3.5	0.5	0.15	4.0	2.4	1.70
Crown proj. field, summer (%)	60.8	11.7	0.19	75.9	44.2	1.72
Crown proj. field, winter (%)	17.1	4.1	0.24	24.2	12.1	2.01
Crown proj. field, annual (%)	39.0	7.6	0.19	49.7	28.1	1.77
Crown proj. image, summer (%)	83.0	5.5	0.07	88.2	66.3	1.33
Crown proj. image, winter (%)	39.4	6.0	0.15	49.5	30.0	1.65
Crown proj. image, annual (%)	61.2	4.8	0.08	67.1	48.1	1.40
Crown volume ($m^3.ha^{-1}$)	34913	11084	0.32	53790	20255	2.66
Crown depth (m)	13.5	3.8	0.28	18.8	5.7	3.31

x = mean, sd = standard deviation, cv = coefficient of variation
[a] For z_o, *LAI* and crown projection of oak, summer, winter and annual mean values are presented.

Douglas fir stands were characterised by relatively large roughness lengths and crown projections compared to those of Scots pine and oak stands (Table 7.7). Roughness lengths of Scots pine and oak stands were not significantly different. Annual mean *LAI* values of oak stands were significantly smaller compared to those of Douglas fir and Scots pine stands. Leaf areas of Douglas fir and Scots pine stands were not significantly different. Scots pine stands were characterised by significantly smaller crown projections, crown depths, crown volumes, and silhouette areas compared to Douglas fir and oak stands. In contrast to Scots pine, oak revealed relatively large crown depths due to the frequent occurrence of stem shoots.

Table 7.8 summarises dimensions of Douglas fir and Scots pine needles. Douglas fir needles can be characterised as short and flat, whereas Scots pine needles were found to be long and (half-) cylinder shaped. Compared to Scots pine needles, the diameter of Douglas fir needles was large, while their mean total surface area was small. No hairs were present on the needles of either species. Considering these findings, it was assumed that the efficiency of Douglas fir needles to collect particles and cloud droplets was more-or-less equal to that of Scots pine needles. The collecting efficiency of oak leaves was assumed to be small compared to that of Douglas fir and Scots pine needles.

TABLE 7.8 Mean dimensions of needles from Douglas fir and Scots pine trees, with *SD* in parentheses (n = 150)

	Douglas fir	Scots Pine
Shape	flat	(half-)cylinder
Diameter (cm)	0.112 (0.013)	0.054 (0.011) - 0.117 (0.019)*
Length (cm)	2.038 (0.309)	5.384 (1.218)
Surface area (cm^2)	0.458 (0.097)	1.610 (0.539)

* Minimum and maximum diameter of the cylinder

Relationships between dry deposition and canopy structure

Relationships between net throughfall and canopy structure characteristics

Pearson's correlation coefficients between annual net throughfall fluxes and canopy structure characteristics of the 30 forest stands are presented in Table 7.9. Net throughfall fluxes of SO_4^{2-}, NO_3^-, NH_4^+ and H^+ were found to relate well with parameters reflecting the aerodynamic roughness as well as collecting surface area of the canopy. For SO_4^{2-}, NO_3^- and NH_4^+, high correlation coefficients were observed with the roughness length of the canopy and for NO_3^- and NH_4^+, also with leaf area. The dependency of SO_4^{2-} and NH_4^+ net throughfall on the roughness length may be explained in the Netherlands by the small surface resistance of SO_2 and NH_3 (see Chapter 4). Dry deposition of these compounds is mainly controlled by the aerodynamic roughness of the canopy surface. Large leaf areas contribute to small aerodynamic resistances. For NO_3^-, the strong dependency on *LAI* and roughness length may be explained by assuming that the surface resistance of NO_2 was inversely related to the leaf area of the forest stand, and/or (in the case of irreversible canopy uptake of NO_2) by considering net throughfall of NO_3^- to be controlled to a large extent by dry deposition of

HNO_3 and NO_3^- aerosol. Dry deposition of HNO_3 compounds is believed to be controlled by atmospheric transfer to the receptor surface (see Chapter 4) and hence by the aerodynamic roughness (length) of the canopy. Dry deposition velocities of NO_3^- above forests are also found to be determined by aerodynamic roughness of the surface, for very rough surfaces (see section 7.3). Net throughfall of H^+ is to a large extent controlled by dry deposition of acids, such as HNO_3 and H_2SO_4, as well as by the ability of the canopy to retain protons. Dry deposition velocities of acids will be determined to a large extent by the aerodynamic properties of the canopy, whereas the ability of the canopy to retain protons will be influenced, for instance, by its total surface area (e.g. nutrient status of the leaves).

TABLE 7.9 Correlation matrix between net throughfall fluxes and canopy structure characteristics for the 30 forest stands

	SO_4^{2-}	NO_3^-	NH_4^+	H^+	Na^+	Cl^-	Mg^{2+}	Ca^{2+}	K^+	HCO_3^-	PO_4^{3-}	mm
z_o	*0.76*	*0.73*	*0.77*	*-0.63*	*0.55*	*0.54*	0.45	*0.65*	-	-	-0.39	*-0.72*
Tree height	*0.64*	0.44	*0.51*	*-0.52*	0.43	0.40	*0.52*	0.64	-	-	-	-0.42
Silh. area	*0.56*	-	-	*-0.50*	-	-	-	-	*0.56*	0.44	-	-0.40
Stem density	-	-	-	-	-	-	-	-	-	-	-	-
LAI	-	*0.78*	*0.73*	*-0.50*	*0.56*	*0.56*	-	0.38	*-0.59*	*-0.62*	*-0.82*	*-0.72*
Cr. pr. field	*0.51*	*0.47*	*0.46*	*-0.72*	-	-	-	-	-	-	-	*-0.55*
Cr. pr. image	*0.52*	*0.56*	*0.56*	*-0.86*	-	-	-	-	-	-	-	*-0.74*
Cr. volume	*0.66*	*0.49*	*0.52*	*-0.64*	-	-	0.37	0.40	-	-	-	*-0.52*
Crown depth	-	-	-	-	-	-	0.40	-	*0.85*	*0.81*	*0.72*	-

Figures in italics indicate correlation coefficients with $p<0.01$.
Correlation coefficients with $p>0.05$ were excluded from the table.

Compared to acidifying compounds, net throughfall of Na^+ and Cl^- showed weaker but still significant relationships with the roughness length and leaf area of the canopy. Next to aerodynamic properties and the total collecting surface area of the canopy, collecting efficiency of individual canopy elements and in-canopy wind speeds also determine the amount of dry deposition of sea salt particles (see section 7.3). Because net throughfall of K^+, HCO_3^- and PO_4^{3-} is mainly controlled by canopy leaching, positive relationships with parameters reflecting the canopy collecting surface area were expected. Such relationships were indeed observed with mean crown depth, but at the same time strong negative relationships were recorded with leaf area. Relationships between net throughfall of K^+, HCO_3^- and PO_4^{3-}, and canopy structure characteristics, were strongly influenced by extremely large fluxes observed in the oak stands and, for this reason, should be interpreted with caution. Tree species seems to be the major controlling factor for these fluxes.

Non-linear regression models were not any better able to explain a larger fraction of the variation in net throughfall than linear regression models. Often an unimodal relationship between dry deposition and *LAI* has been suggested in the literature (Meyers *et al.*, 1989; Lovett and Reiners, 1986; Ivens, 1990), but in this study relationships between net throughfall and *LAI* turned out to be linear in the *LAI* range of 2.4 to 12.0. Relationships between net throughfall and other canopy structure characteristics were also found to be linear.

Relationships between net throughfall (corrected for canopy exchange) and canopy structure characteristics
To obtain more insight into the relation between dry deposition and canopy structure, net throughfall was corrected for the contribution of canopy exchange by using the Ulrich (1993) and Van der Maas and Pape (1991) model before relating them to the canopy structure characteristics. Correlation coefficients between corrected net throughfall fluxes of SO_4^{2-}, NO_3^-, Na^+, Cl^-, Mg^{2+}, Ca^{2+}, K^+, and PO_4^{3-} on the one hand, and canopy structure characteristics on the other, were found equal to those computed with uncorrected net throughfall fluxes (Table 7.9). For Cl^-, Mg^{2+}, Ca^{2+}, K^+, and PO_4^{3-}, this can be attributed to the method used in the model to compute the contribution of canopy exchange to net throughfall, and for SO_4^{2-}, NO_3^- and Na^+ to the assumption that canopy exchange is negligible (Draaijers *et al.*, 1994). Corrected net throughfall fluxes of NH_4^+ related fairly well with roughness length (R=0.71; p<0.001), LAI (R=0.66; p<0.001) and crown projection estimated from scanned images (R=0.71; p<0.001). Corrected net throughfall fluxes of H^+ were found to relate well with LAI (R=-0.85; p<0.001) and crown projection estimated from scanned images (R=-0.75; p<0.001).

Impact of season on relationships between net throughfall and canopy structure characteristics
Relationships between net throughfall of SO_4^{2-}, NO_3^- and NH_4^+, and canopy structure characteristics, were strongly influenced by season. Stronger relationships were observed in the winter period, which may be attributed to the relatively low physiological activity at this time, giving canopy exchange processes minor influence on the composition of throughfall. More than 70% of the variation in winter net throughfall of SO_4^{2-}, NO_3^- and NH_4^+ could be explained by differences in aerodynamic roughness length of the canopy (Figure 7.8 - 7.10) but strong relationships were also observed with *LAI* (R=0.59, 0.77 and 0.76, respectively), crown projection estimated in the field (R=0.69, 0.78 and 0.76, respectively) and crown projection estimated from scanned images (R=0.64, 0.75 and 0.74, respectively).

Relationships between net throughfall and multiple canopy structure characteristics
Multiple regression was used to see if multiple canopy structure characteristics regulated annual net throughfall. Strong interrelationships were found between most canopy structure characteristics (Table 7.10). The roughness length, for example, related significantly to tree height, silhouette area, leaf area, crown projection and crown volume. To determine whether combinations of two likely factors explain a larger amount of the variance in net throughfall than single factors, multiple regression was performed using all possible pairs of independent variables. Roughness length and crown depth were found to explain large parts of the variance in net throughfall of NO_3^- (R=0.84), NH_4^+ (R=0.83), Na^+ (R=0.62) and Cl^- (R=0.61). Similar correlation coefficients were obtained using tree height and leaf area as explanatory variables. Other combinations of two independent canopy structure variables were not able to explain a larger amount of the variance than single variables.

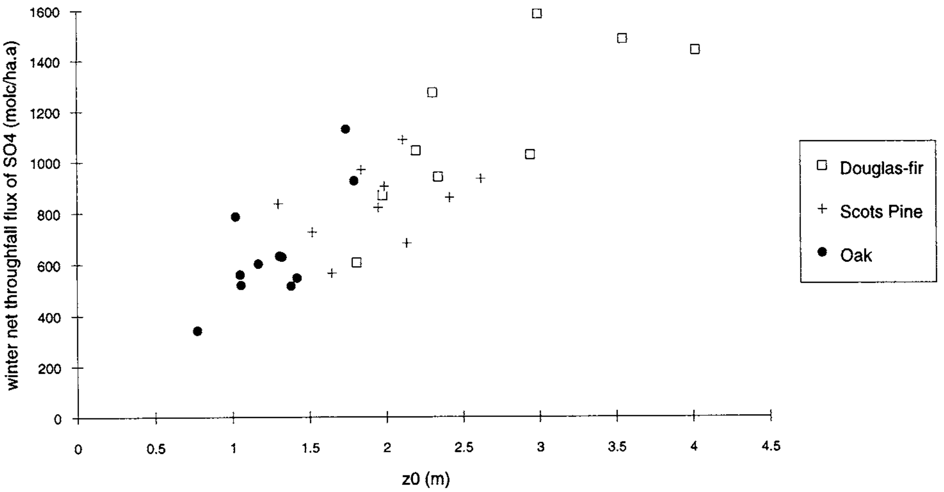


FIGURE 7.8 Relationship between winter net throughfall fluxes of SO_4^{2-} and the aerodynamic roughness length of the canopy. Douglas fir is indicated by squares, Scots pine by plus signs and oak by dots. The following relationship is valid: $[NTF\text{-}\ SO_4^{2-}]=344.9[z_o]+201.3$ (R=0.84; p<0.001).

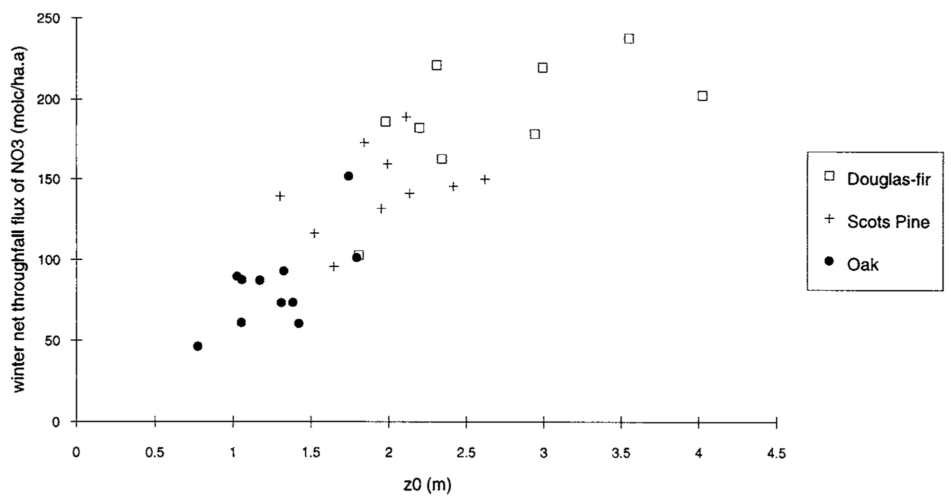


FIGURE 7.9 Relationship between winter net throughfall fluxes of NO_3^- and the aerodynamic roughness length of the canopy. Douglas fir is indicated by squares, Scots pine by plus signs and oak by dots. The following relationship is valid: $[NTF\text{-}NO_3^-]=59.6[z_o]+21.5$ (R=0.84; p<0.001).

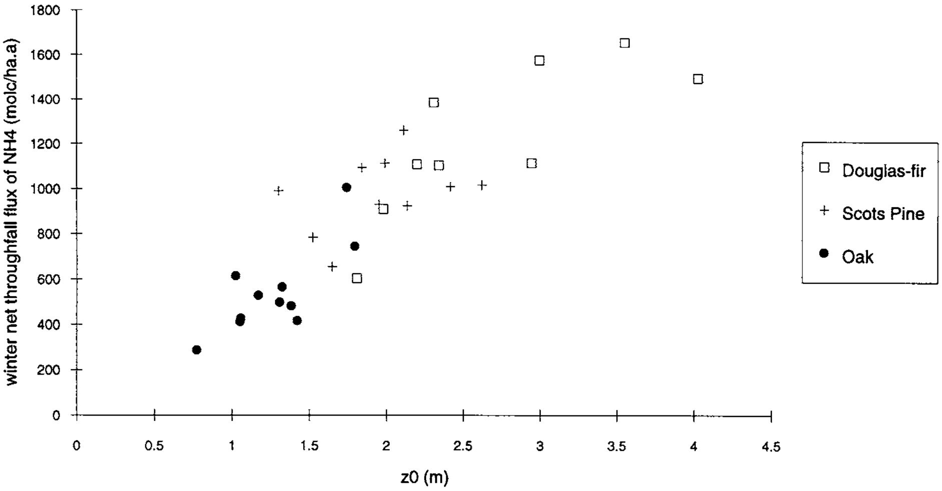


FIGURE 7.10 Relationship between winter net throughfall fluxes of NH_4^+ and the aerodynamic roughness length of the canopy. Douglas fir is indicated by squares, Scots pine by plus signs and oak by dots. The following relationship is valid: $[NTF\text{-}NH_4^+]=431.0[z_o]+65.5$ (R=0.88; p<0.001).

TABLE 7.10 Interrelationships between the individual canopy structure characteristics of the 30 forest stands

	z_0	Tree height	Silhouette area	Stem density	*LAI*	Crown proj., field	Crown proj., image	Crown volume	Crown depth
z_0	*1.00*	*0.63*	*0.63*	--	*0.55*	0.45	*0.59*	*0.68*	--
Tree height		*1.00*	0.39	*-0.50*	--	*0.52*	--	*0.60*	0.44
Silhouette area			*1.00*	--	--	*0.58*	*0.60*	*0.79*	*0.61*
Stem density				*1.00*	--	--	--	--	--
LAI					*1.00*	--	0.46	--	*-0.64*
Crown proj., field						*1.00*	*0.79*	*0.85*	--
Crown proj., image							*1.00*	*0.67*	--
Crown volume								*1.00*	--
Crown depth									*1.00*

Italics indicate correlation coefficients with p<0.01.

Correlation coefficients with p>0.05 were not considered significant and thus were excluded from the table.

Relationships between net throughfall and canopy structure characteristics for single tree species

To avoid differences in surface resistance, collecting efficiency, canopy exchange and uncertainties in drag coefficients, relationships between annual net throughfall and canopy structure characteristics were also examined for single tree species. Essentially, similar relationships were found between net throughfall and canopy structure characteristics to those observed considering all the 30 stands together (Table 7.10). However, many relationships were somewhat weaker, which may be attributed to the limited range in canopy structure characteristics observed within single tree species. For example, for all the stands collectively the ratio between maximum and minimum roughness length was 3.62 (Table 7.6), whereas within single tree species this ratio was only 1.85-2.23 (Table 7.7). For *LAI*, the difference was even more extreme. Whereas the ratio between maximum and minimum *LAI* was 5.00 for all the stands collectively, it was only 1.57-1.70 within single tree species.

Predicting net throughfall using simple regression models with canopy structure and information on pollution climate

To see if it was possible to predict net throughfall fluxes using simple linear regression models with canopy structure characteristics, winter net throughfall fluxes of SO_4^{2-}, NO_3^- and NH_4^+ for each of the 30 stands were predicted by using regression equations with a roughness length based on the data of the remaining 29 stands. Predicted and measured fluxes are presented in Figures 7.11-13. Reasonable agreement was observed (R=0.60, 0.50 and 0.58 for SO_4^{2-}, NO_3^- and NH_4^+, respectively) although the regression models were found to overestimate net throughfall fluxes at low values and tended to underestimation at higher values. Using annual net throughfall fluxes and/or other canopy structure characteristics (e.g. *LAI* or crown projection) as a basis for the regression models yielded similar conclusions, though differences between predicted and measured net throughfall fluxes were larger.

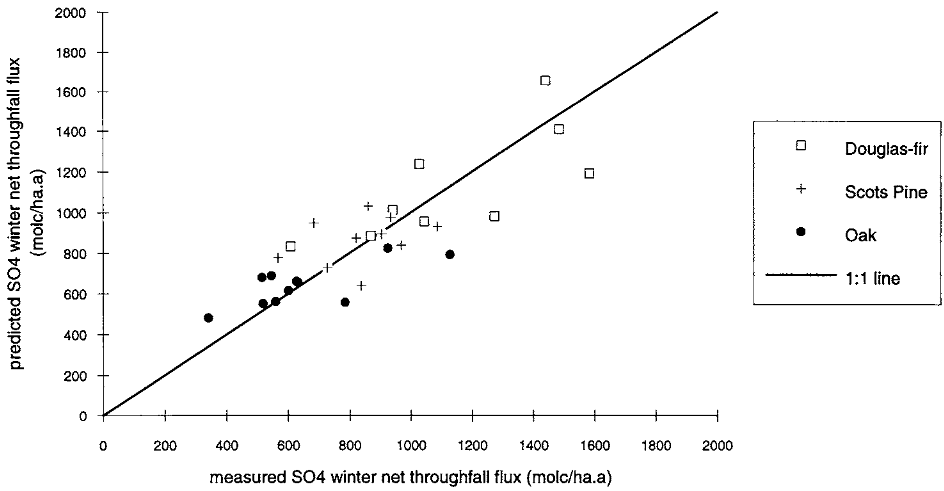


FIGURE 7.11 Winter net throughfall fluxes of SO_4^{2-} predicted from linear regression models with the roughness length of the canopy, compared to measured winter net throughfall fluxes. Douglas fir is indicated by squares, Scots pine by plus signs and oak by dots.

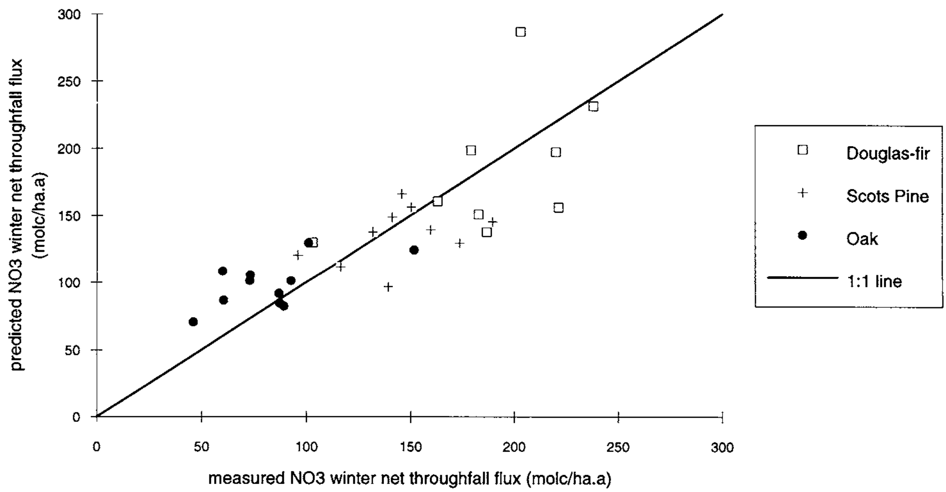


FIGURE 7.12 Winter net throughfall fluxes of NO_3^- predicted from linear regression models with the roughness length of the canopy, compared to measured winter net throughfall fluxes. Douglas fir is indicated by squares, Scots pine by plus signs and oak by dots.

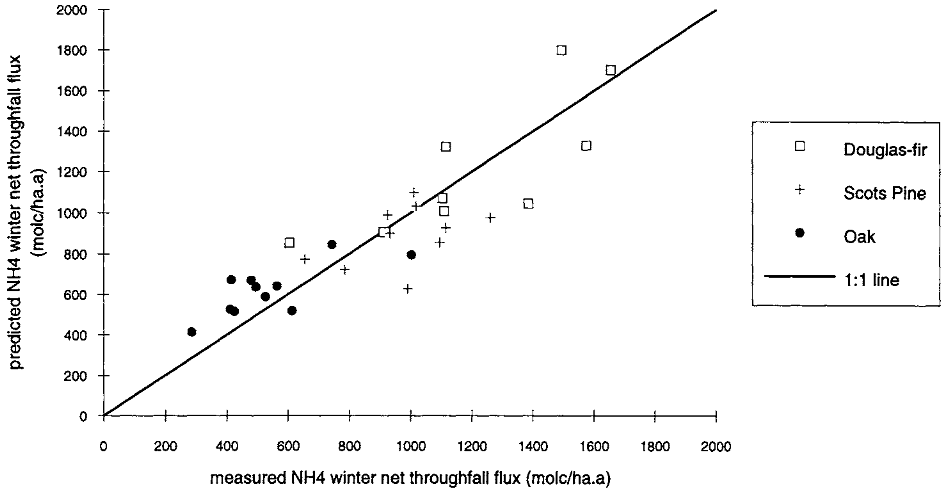


FIGURE 7.13 Winter net throughfall fluxes of NH_4^+ predicted from linear regression models with the roughness length of the canopy, compared to measured winter net throughfall fluxes. Douglas fir is indicated by squares, Scots pine by plus signs and oak by dots.

7.2.4 THE IMPACT OF FOREST EDGES: RESULTS AND DISCUSSION

Evaluation of canopy structure parametrisation

Table 7.11 presents mean canopy and edge structure data of the eight forest edges. Stem density, *LAI*, leaf-area density and silhouette area density were found to differ greatly among the edges. Relatively large values for these structure characteristics were recorded for location F, and to a lesser extent for location C. Forest edges consisting of European larch trees (locations A, E and G) have relatively large crown depths as a result of the frequent occurrence of stem shoots. The roughness length of the upwind terrain as estimated from the terrain classification of Wieringa (1992) was found to range between 0.03 m and 0.08 m, indicating rather 'smooth' conditions of the upwind terrain for all edges. The roughness length of the forest canopy as estimated from the geometrical model of Lettau (1969) has no physical meaning because the Lettau model only yields roughness lengths comparable to micrometeorologically derived roughness lengths for homogeneous canopy surfaces with an 'infinite' length.

Within an individual forest edge, canopy and edge structure characteristics showed only small spatial variability. Coefficients of variation were nearly always less than 0.20 (not shown in Table 7.11). For location A, the tree height was found to increase from 16 m to approximately

24 m within the first two edge heights. Tree height increment with distance from the edge was less, though still significant for location E (12 m to 15 m) and location G (13 m to 17 m). Only European larch stands showed such an increment. Probably, European Larch trees are probably relatively susceptible to reduced growth as a result of wind exposure. Other canopy or edge structure characteristics did not show significant gradients with distance from the edge. At the very edge of the forest edges A, B, C, F and H, relatively dense understorey vegetation (shrubs and/or small trees) was present.

TABLE 7.11 Canopy and edge structure characteristics of the eight forest edges, showing averages of 7 or 8 recordings (structure characteristics were determined around each throughfall gutter) with the exception of porosity and the roughness length of the upwind terrain

Forest edge[a]:	A	B	C	D	E	F	G	H
Canopy structure characteristics								
z_o, canopy (m)	1.6	1.3	1.4	1.0	1.8	2.4	1.3	1.3
Tree height (m)	22.3	12.7	9.0	13.7	14.1	9.9	16.0	13.7
Silhouette area (m^2 ha^{-1})	8771	8351	13453	5432	15145	25124	10413	7348
Stem density (ha^{-1})	630	1264	2490	531	830	2533	329	653
LAI (-)	2.8	6.9	8.8	5.4	2.3	15.7	2.6	5.2
Crown proj., field (%)	21.3	18.0	29.6	17.7	12.7	49.2	15.5	23.0
Crown proj., image (%)	63.6	65.3	67.7	59.3	62.1	87.2	60.7	58.3
Crown volume (m^3 ha^{-1})	33159	13763	18847	10775	26721	35489	30133	16260
Crown depth (m)	13.1	3.4	4.1	4.2	7.2	6.6	7.8	4.3
Edge structure characteristics								
Relative height (m)	21.5	12.6	8.5	13.0	13.8	9.7	15.3	13.0
Porosity, 1 edge height (%)	39.3	41.8	29.7	57.5	45.8	43.3	45.5	45.0
Porosity, 2 edge heights (%)	36.3	34.3	28.8	42.3	39.4	15.4	43.6	33.7
Stand leaf area d. (m^2 m^{-3})	0.13	0.54	0.98	0.40	0.16	1.58	0.16	0.38
Stand silh. area d. (m^2 m^{-3})	0.04	0.07	0.15	0.04	0.11	0.26	0.07	0.05
Crown leaf area d. (m^2 m^{-3})	0.22	2.03	2.14	1.27	0.32	2.38	0.33	1.21
Crown silh. area d. (m^2 m^{-3})	0.07	0.25	0.33	0.13	0.21	0.38	0.13	0.17
z_o, upwind terrain (m)	0.08	0.03	0.07	0.08	0.04	0.03	0.08	0.08

[a]Tree species: *Larix decidua* for forest edge A, E and G; *Pinus sylvestris* for forest edge B, D and H. *Pinus nigra* for forest edge C and *Picea abies* for forest edge F.

For z_o 'canopy', *LAI*, crown projection, relative height, porosity, leaf area density and z_o 'upwind terrain', annual mean values are presented which were calculated by averaging summer and winter values.

Dry deposition gradients in the forest edges
Horizontal net throughfall flux gradients for the eight forest edges are presented in Figure 7.14. In general, an exponential increase with decreasing distance to the edge was found, with in most cases a considerable scatter around the curve. To a large extent this may be the result of non-representative throughfall sampling as net throughfall at each distance from the edge was measured by means of only one fixed throughfall gutter. Because of this the mean flux at that distance may be underestimated or overestimated by 32%-45%. depending on the ion under consideration (Van Ek and Draaijers, 1991). The observed scatter could not be explained by the variability in canopy structure characteristics in the edge. From Figure 7.14 it is also clear that the width of the zone with enhanced net throughfall fluxes was usually five edge heights at most. This is in agreement with throughfall studies performed in other forest edges (Hasselrot and Grennfelt, 1987; Draaijers *et al.*, 1988; Beier and Gundersen, 1989).

A schematical presentation of a mean net throughfall flux gradient in a forest edge is shown in Figure 7.15. Discrimination is made here between basic net throughfall and edge-net throughfall. The basic net throughfall is defined as the net throughfall flux which would have been measured without the edge effect; edge-net throughfall is defined as the net throughfall flux resulting from the edge effect. Using the data from Figure 7.14, edge net throughfall fluxes were computed by calculating best-fitting decay curves using a power-law relationship ($NTF=a(x/h)^b$ in which NTF = net throughfall flux, x/h = distance to forest edge divided by edge height, with a and b as regression coefficients), and subsequent computing of the area under this curve through integration. Edge net throughfall fluxes were determined between x/h=0.25 and x/h=5.0. The zone between x/h=0 and x/h=0.25 was not included because net throughfall fluxes in this zone were not measured in the field, and infinitely high edge net throughfall fluxes would be obtained, otherwise (at x/h=0, NTF equals ∞ for b<0). This area could be included by fitting other curves (for example, $NTF=a(e^{-x/h})+b$), but then usually considerable smaller amounts of variance were explained. For this reason, power-law decay curves were preferred. Basic net throughfall in the zone between x/h=0.25 and x/h=5 was assumed equal to the net throughfall flux at x/h=5 multiplied by 4.75 (= 5 minus 0.25). Dividing edge + basic net throughfall by basic net throughfall yielded whole-edge integrated net throughfall enhancement factors (*WEINTE* factors). If no significant power-law decay curve was found, edge net throughfall fluxes were assumed to be zero, and the *WEINTE* factor equal to 1.

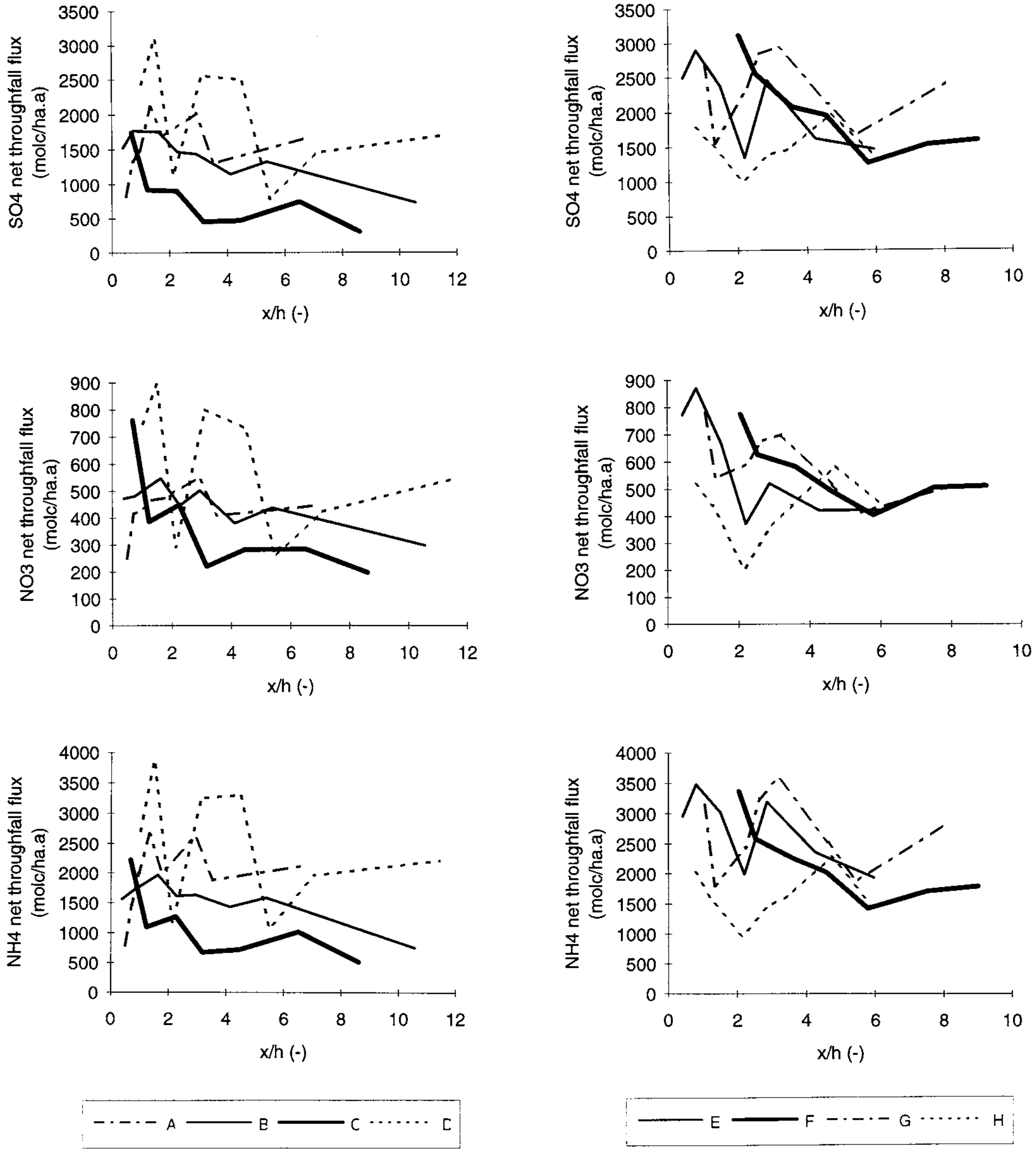


FIGURE 7.14 Net throughfall flux gradients of SO_4^{2-}, NO_3^- and NH_4^+ for the eight forest edges. Distance to edge divided by edge height is *x/h*

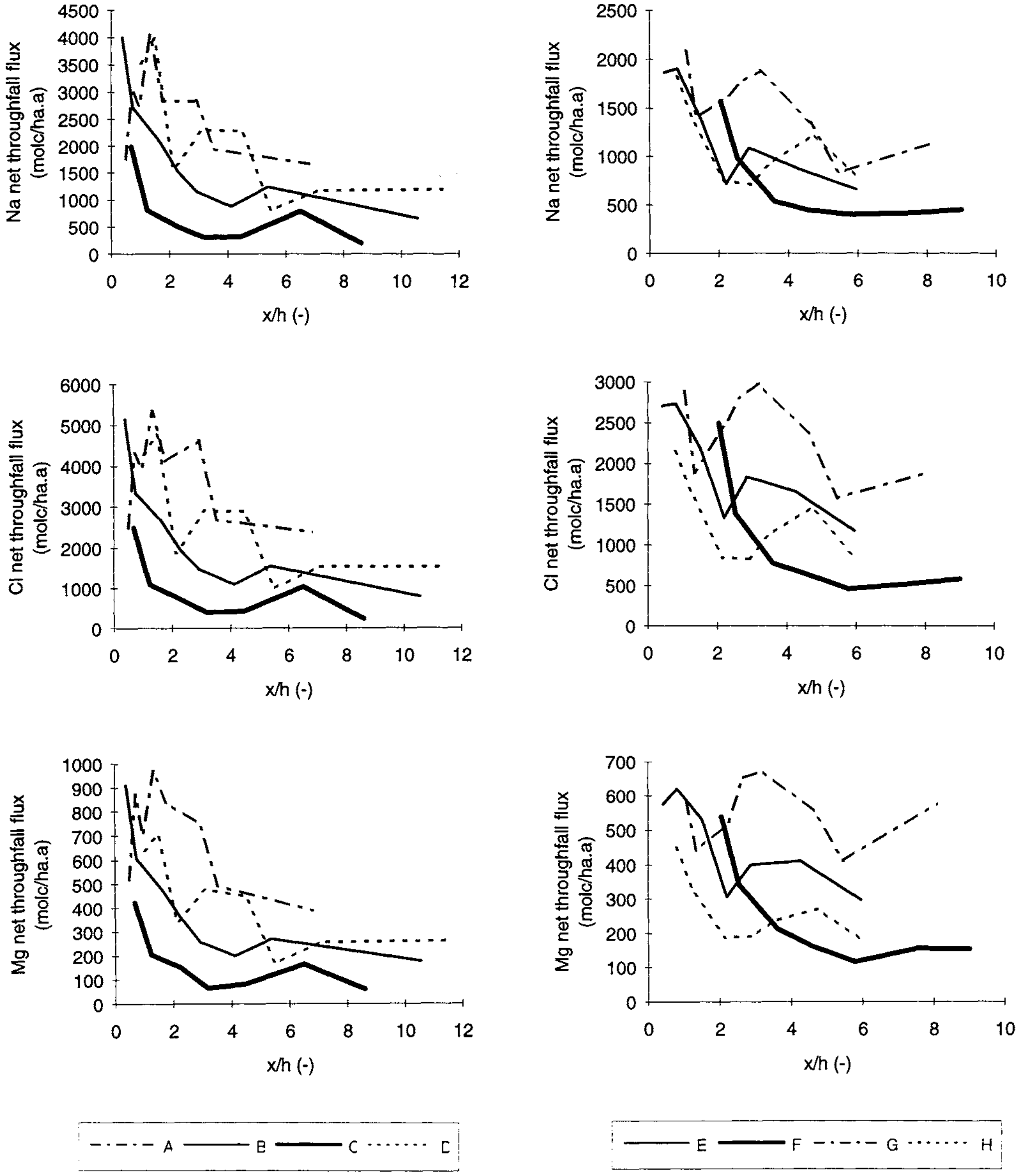


FIGURE 7.14 *(continued)* Net throughfall flux gradients of Na^+, Cl^- and Mg^{2+} for the eight forest edges. Distance to edge divided by edge height is *x/h*

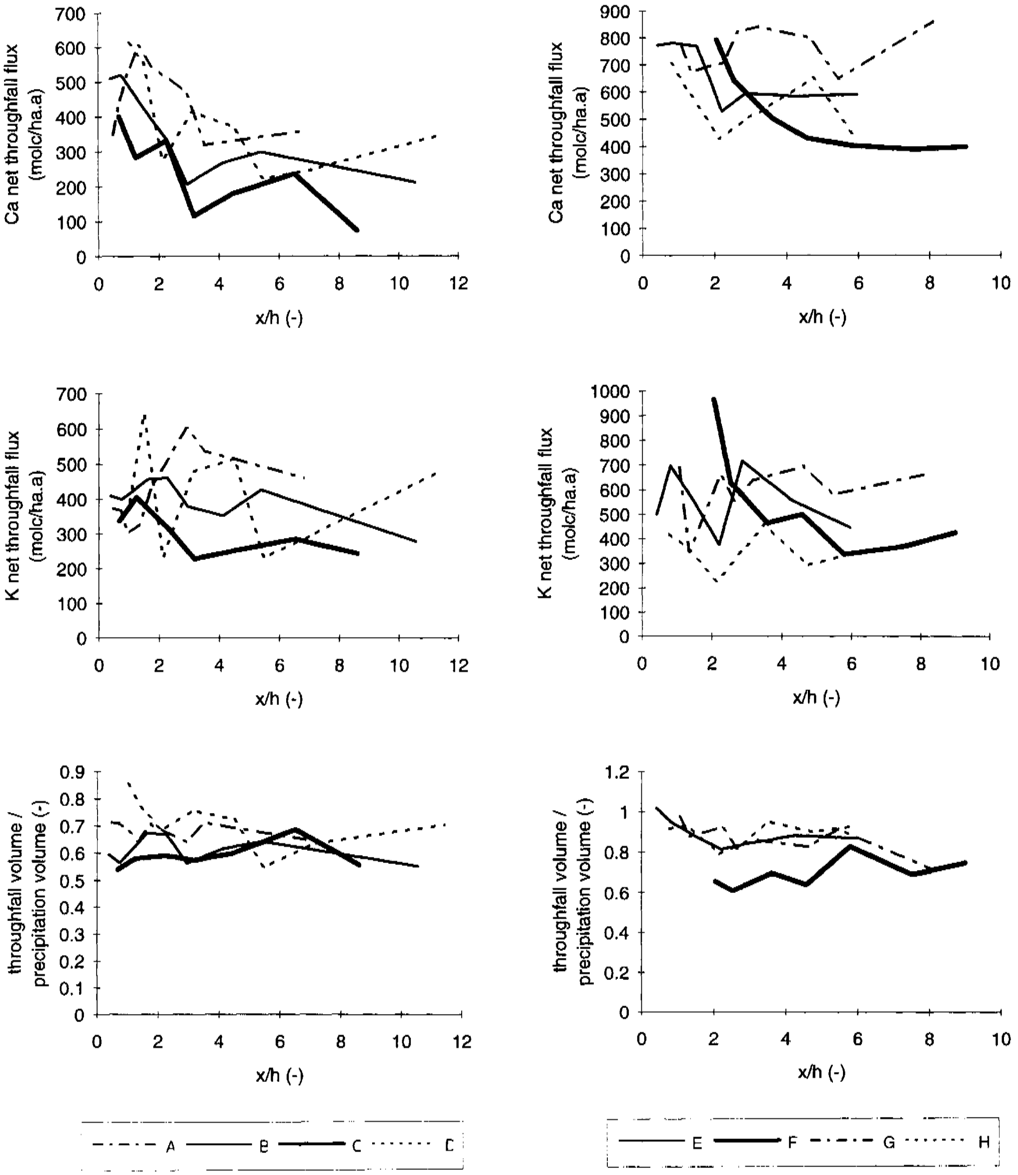


FIGURE 7.14 *(continued)* Net throughfall flux gradients of Ca^{2+} and K^{+}, and gradients of the ratio between throughfall volume and precipitation volume for the eight forest edges. Distance to edge divided by edge height is *x/h*

WEINTE factors for the eight forest edges are shown in Table 7.12. A significant net throughfall enhancement was observed for Na^+, Cl^- and Mg^{2+} in all edges. Net throughfall fluxes at location A also decreased with distance from the edge but the decrease only started at approximately two edge heights from the forest edge. A gradually increasing tree height with distance from the edge (16 up to 24 m) and the dense understorey vegetation present at the very edge of this forest may have resulted in a smooth roughness transition zone. Air entering such a roughness transition will tend to be lifted over, rather than to penetrate into the forest edge. It must be noted, however, that the net throughfall pattern observed for location A and its large deviation from the 'ideal' power-law decay curve may also be explained by non-representative throughfall sampling.

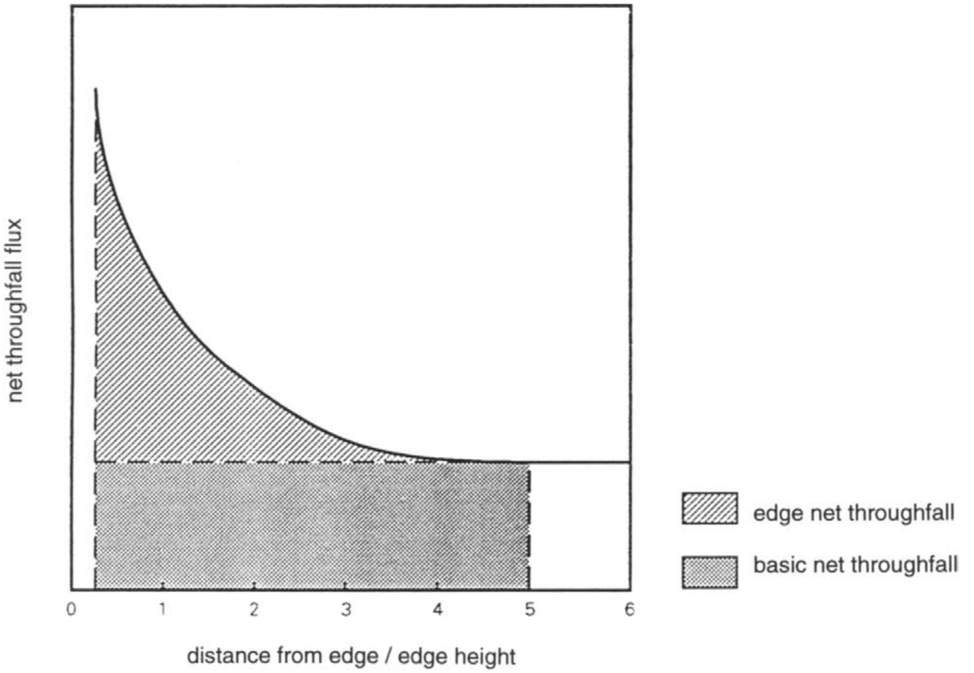


FIGURE 7.15 'Ideal' net throughfall flux gradient in a forest edge.

The *WEINTE* factors for SO_4^{2-}, NO_3^- and NH_4^+ were considerably smaller than those for Na^+, Cl^- and Mg^{2+} (Table 7.12). This may be attributed to the different sources for these ions in throughfall. Net throughfall of SO_4^{2-}, NO_3^- and NH_4^+ is controlled to a large extent by dry deposition of gases and fine (sub-micron) particles, whereas net throughfall of Na^+, Cl^- and Mg^{2+} is mainly controlled by dry deposition of coarse sea salt particles. Deposition of coarse particles is very much enhanced in forest edges as a result of high in-canopy wind speeds, which enhances impaction efficiencies considerably. Edge effects seem to have only minor impact on the deposition of gases and fine particles. For the locations A, D, G and H, net throughfall fluxes of SO_4^{2-}, NO_3^- and NH_4^+ were not significantly enhanced.

TABLE 7.12 Whole-edge integrated net throughfall enhancement (*WEINTE*) factors for the eight forest edges (x = mean of all edges)

	A	B	C	D	E	F	G	H	x
SO_4^{2-}	1.00	1.20	1.73	1.00	1.23	1.64	1.00	1.00	1.23
NO_3^-	1.00	1.11	1.54	1.00	1.31	1.31	1.18	1.00	1.18
NH_4^+	1.00	1.16	1.54	1.00	1.15	1.55	1.00	1.00	1.18
Na^+	1.00	1.71	2.00	1.73	1.48	2.50	1.32	1.35	1.64
Cl^-	1.00	1.74	2.07	1.66	1.33	3.11	1.00	1.39	1.66
Mg^{2+}	1.00	1.66	1.88	1.58	1.26	2.57	1.00	1.39	1.54
Ca^{2+}	1.00	1.31	1.65	1.33	1.13	1.58	1.00	1.00	1.25
K^+	0.82	1.00	1.16	1.00	1.00	1.74	1.00	1.00	1.09
Volume	1.00	1.00	1.00	1.09	1.05	1.00	1.09	1.00	1.03

Although Ca^{2+} and K^+ net throughfall fluxes were also likely to be influenced by dry deposition of coarse particles (e.g. soil dust, pollen), *WEINTE* factors for these ions were much lower compared to those of Na^+, Cl^- and Mg^{2+} (Table 7.12). This was a surprising result, given the fact that these fluxes are probably strongly influenced by canopy leaching (Van Ek and Draaijers, 1993). Canopy leaching was assumed to be relatively intense in forest edges due to vigorous transpiration of trees in wind-exposed forest edges through which the supply of Ca^{2+} and K^+ from the soil to the foliage was expected to be relatively high. However, the nutrient status of trees at the forest border may have been lowered as a result of extensive leaching of Ca^{2+} and K^+ out of the topsoil due to the high acid inputs of the last decades. The significantly reduced K^+ net throughfall fluxes measured near the edge of location A may be an indication of this. For most other locations, this reduced leaching at the edge apparently counterbalanced enhanced dry deposition of K^+-containing particles, resulting in no net throughfall flux enhancement. Only for locations C and F were significantly enhanced net throughfall fluxes of K^+ observed. These locations also showed the highest *WEINTE* factors for Na^+, Cl^- and Mg^{2+}.

For the locations D, E and G, throughfall volumes were slightly, though significantly, enhanced (Table 7.12). For location E, throughfall volumes very close to the edge were even found to exceed precipitation volumes. Enhanced throughfall volumes near the forest edge may to a large extent be attributed to the horizontal influx of rain droplets through the trunk area. In the trunk area of the forest edges D, E and G, no understorey vegetation taller than 1 m was present and the height of the living crown base exceeded 7 m. Other locations showed relatively high and dense understory vegetation near the very edge of the forest, limiting the horizontal influx of rain droplets. Relatively large turbulence intensities in forest edges, through which interception and capture of (small) rain droplets and cloud and fog water droplets is efficient (Weathers *et al.*, 1992), may also have contributed to the enhanced throughfall volumes observed in some forest edges.

On average, ratios of net throughfall fluxes at x/h=0.25 and x/h=5 (Table 7.13) were similar to those found by Potts (1978), Hasselrot and Grennfelt (1987), Draaijers *et al.* (1988) and Beier and Gundersen (1989). At the very edge of the forest, net throughfall fluxes of Na^+, Cl^- and Mg^{2+} were, on average, approximately five times larger than net throughfall fluxes in the interior forest. For the locations C and F extremely large ratios were computed, where net throughfall fluxes of Cl^- at x/h=0.25 was up to 20 times larger than those at x/h=5.0. Net throughfall fluxes of SO_4^{2-}, NO_3^- and NH_4^+ were, on average, doubled at the very edge of the forest.

TABLE 7.13 Ratio between the net throughfall flux at x/h = 0.25 and that at *x/h* = 5 for the eight forest edges

	A	B	C	D	E	F	G	H	*x*
SO_4^{2-}	1.00	1.86	5.18	1.00	1.98	4.53	1.00	1.00	2.19
NO_3^-	1.00	1.43	3.83	1.00	2.42	2.41	1.76	1.00	1.86
NH_4^+	1.00	1.63	3.78	1.00	1.63	3.89	1.00	1.00	1.87
Na^+	1.00	5.06	7.44	5.15	3.45	12.31	2.44	2.64	4.94
Cl^-	1.00	5.26	7.98	4.67	2.53	19.39	1.00	2.85	5.59
Mg^{2+}	1.00	4.64	6.42	4.07	2.16	13.09	1.00	2.87	4.41
Ca^{2+}	1.00	2.42	4.55	2.50	1.51	4.11	1.00	1.00	2.26
K^+	0.47	1.00	1.64	1.00	1.00	5.22	1.00	1.00	1.54
Volume	1.00	1.00	1.00	1.36	1.20	1.00	1.33	1.00	1.11

x/h = distance to edge divided by edge height; *x* represents the mean of all edges.

Impact of canopy/edge structure and edge aspect

Pearson's correlation coefficients between *WEINTE* factors and canopy/edge structure characteristics are presented in Table 7.14. Enhancement factors correlated positively with parameters, reflecting the density of the canopy/edge. For Na^+, Cl^- and Mg^{2+}, very strong relationships were observed with leaf area and leaf-area density. Such relationships were already predicted by Wiman and Ågren (1985) using a modelling approach and suggest that in-canopy wind speeds determine, to a large extent, differences between forest edge and forest interior dry deposition. *WEINTE* factors of SO_4^{2-}, NO_3^- and NH_4^+ correlated best with silhouette area density and stem density. This suggests that for these ions the differences between forest edge and forest interior dry deposition can be explained by differences in drag forces and, thus, turbulence intensities. Furthermore, strong relationships were found between throughfall volume enhancement factors on the one hand, and crown projection and edge porosity on the other. This is in agreement with the hypothesis that enhanced throughfall volumes were largely the result of horizontal influx of rain droplets.

TABLE 7.14 Correlation matrix between whole-edge integrated net throughfall enhancement (*WEINTE*) factors and canopy/edge structure characteristics for the eight forest edges

	SO_4^{2-}	NO_3^-	NH_4^+	Na^+	Cl^-	Mg^{2+}	Ca^{2+}	K^+	mm
Canopy structure characteristics									
z_o, canopy	-	-	-	-	-	-	-	0.75	-0.81
Tree height	-0.74	-	-0.74	*-0.86*	-0.75	-0.79	-0.80	-	-
Silhouette area	0.75	-	0.79	0.71	0.75	-	-	*0.88*	-0.78
Stem density	*0.98*	0.76	*0.99*	*0.85*	*0.87*	*0.87*	*0.90*	0.78	0.76
LAI	0.77	-	*0.83*	*0.92*	*0.98*	*0.97*	0.81	*0.93*	*-0.86*
Crown proj., field	0.72	-	0.78	0.78	0.77	*0.89*	-	*0.90*	*-0.92*
Crown proj., image	0.75	-	0.81	0.81	*0.89*	*0.85*	-	*0.93*	*-0.91*
Crown volume -	-	-	-	-	-	-	-	-	
Crown depth	-	-	-	-	-	-	-	-	-
Edge structure characteristics									
Relative height -0.73	-	-0.73	*-0.85*	-0.74	-0.78	-0.79	-	-	
Porosity, one edge h.	-	-	-	-	-	-	-	-	-
Porosity, two edge h.	-0.78	-	*-0.83*	*-0.77*	*-0.89*	*-0.87*	-	*-0.87*	*0.97*
Stand leaf area dens.	*0.85*	-	*0.89*	*0.94*	*0.98*	*0.97*	*0.86*	*0.93*	*-0.84*
Stand silh. area dens.	*0.87*	0.71	*0.90*	*0.86*	*0.88*	*0.84*	0.72	*0.94*	*-0.80*
Crown leaf area dens.	0.72	-	0.75	*0.86*	*0.86*	*0.90*	*0.86*	-	-
Crown silh. area dens.	*0.93*	0.76	*0.94*	*0.91*	*0.89*	*0.89*	*0.85*	*0.83*	-0.71
z_o, upwind terrain	-	-	-	-	-	-	-	-	-

Italic values indicate correlation coefficients with $p<0.01$. Correlation coefficients with $p>0.05$ were excluded from the table.

The impact of edge aspect on dry deposition gradients in forest edges could not be assessed directly in this experiment because all edges had the same southwest exposition. However, to obtain some insight on the impact of edge aspect, the percentage to which a particular wind direction contributed to the total pollutant dose and the total air mass supply were computed (Figure 7.16). The pollutant dose of a particular wind direction was calculated by multiplying air mass supply (= mean wind speed multiplied by the duration) and average pollutant concentration during the measurements. Information on air mass supply was gathered from hourly based data on wind direction and wind speed from the meteorological-station 'De Bilt', located approximately 20 km northwest of Leersum. These data were made available by the Royal Netherlands Meteorological Institute (KNMI). Concentrations of SO_2, NO_2, NO gas, and SO_4^{2-}, NO_3^-, NH_4^+ and Cl^- aerosol, were gathered from respective hourly and daily measurements taken by RIVM at Bilthoven (near 'De Bilt'). Air concentrations of Na^+, Mg^{2+}, Ca^{2+} and K^+ were not measured, but gathered from results of an observational study in the western part of the Netherlands on precipitation chemistry as function of the surface wind direction (Weijers and Vugts, 1990). The concentrations were estimated using the concentrations in precipitation and scavenging ratios presented by Eder and Dennis (1990). Unfortunately, no information was available on the percentage a particle wind direction contributed to the total dose of HNO_3, HNO_2, NH_3 and HCl.

It can be observed from Figure 7.16 that southern, southwesteren and westeren wind directions contributed the bulk (50-70%) of the dose of each pollutant. This may be attributed to the fact that these are the prevailing winds in the Netherlands; they also show the largest mean wind speeds. For SO_2, NO_2, NO gas and SO_4^{2-}, NO_3^- and NH_4^+ aerosol, the contribution of winds from the east to the total pollutant dose was also relatively large. This was due to the highly polluted air masses coming from the Ruhr area and Eastern Europe. The contribution of western, northwestern and northern winds was reasonably high for Na^+, Cl^- and Mg^{2+}, due to the supply of air masses which traversed the North Sea and/or the Atlantic Ocean. These air masses are rich in sea salt particles. Compared to the locations E-H, locations A-D showed relatively large percentages of the total pollutant dose contributed by wind directions exposed to the forest edge.

Assuming that dry deposition is only enhanced when the wind enters the forest edge, *WEINTE* factors for edges with other than southwestern aspects can be computed by:

$$WEINTE_x = [DOSEEXP_x/DOSEEXP_{sw} * (WEINTE_{sw}-1)] + 1 \qquad [7.2]$$

in which $WEINTE_x$ is the whole-edge integrated net throughfall enhancement factor for an edge with aspect *x*; $DOSEEXP_x$ is the percentage of the total pollutant dose supplied with wind directions exposed to the forest edge (e.g. for a forest edge with western aspect, $DOSEEXP_w$ is equal to the sum of the percentages for southwestern, western, northwestern, half of the southern and half of the northern wind direction); $DOSEEXP_{sw}$ is the percentage of the total pollutant dose supplied with wind directions exposed to the forest edge with a southwestern aspect; and $WEINTE_{sw}$ equals the whole-edge integrated net throughfall enhancement factor for an edge with a southwestern aspect. The latter was assumed equal to the mean of the forest edges A-H (Table 7.12). For the computation of $DOSEEXP_x$, two yearly averaged percentages for each wind direction were used. Moreover, computed $DOSEEXP_x$ values were weighted to the relative contribution of the pollutant to the net throughfall flux (Draaijers, 1993). For the computation of *WEINTE* factors of NO_3 and NH_4, only wind direction-related doses of NO_2, NO, NO_3^- and NH_4^+ aerosols could be used because data for HNO_3, HNO_2 and NH_3 were lacking. For the computation of whole-edge integrated enhancement factors of throughfall volumes, essentially the same procedure was followed but, instead of the percentage of the total pollutant dose, the percentage of the total air mass supply was used. Besides the percentage of the total pollutant dose supplied with wind directions exposed to the edge, the actual dry deposition in forest edges is also dependent on the 'deposition climate'. The latter will most probably change with edge exposition but these changes are not considered in the calculation of $WEINTE_x$ factors.

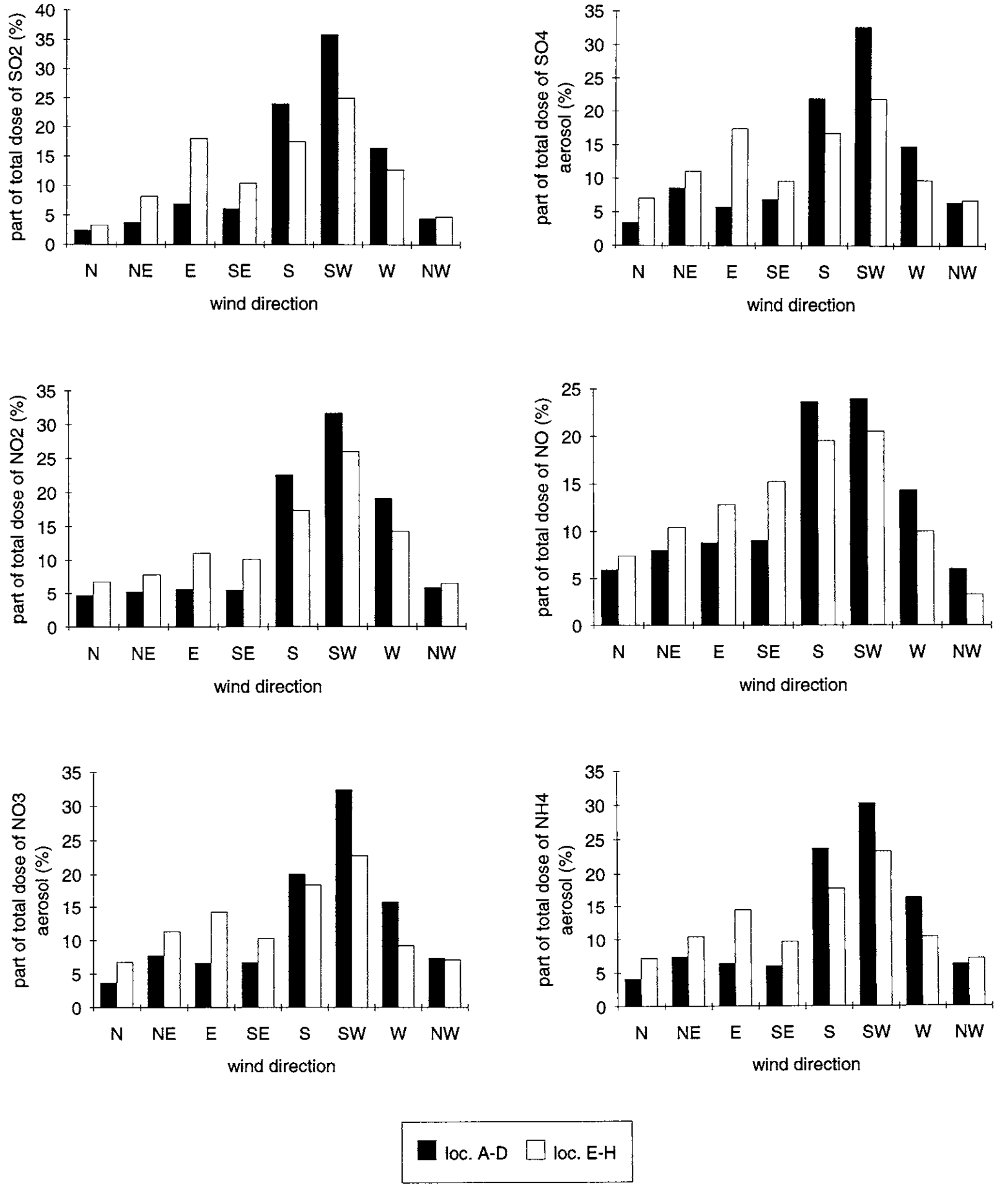


FIGURE 7.16 The percentage to which a particular wind direction contributed to the total pollutant dose of SO_2, SO_4^{2-}, NO_2, NO, NO_3^-, and NH_4^+ during the measurements, respectively

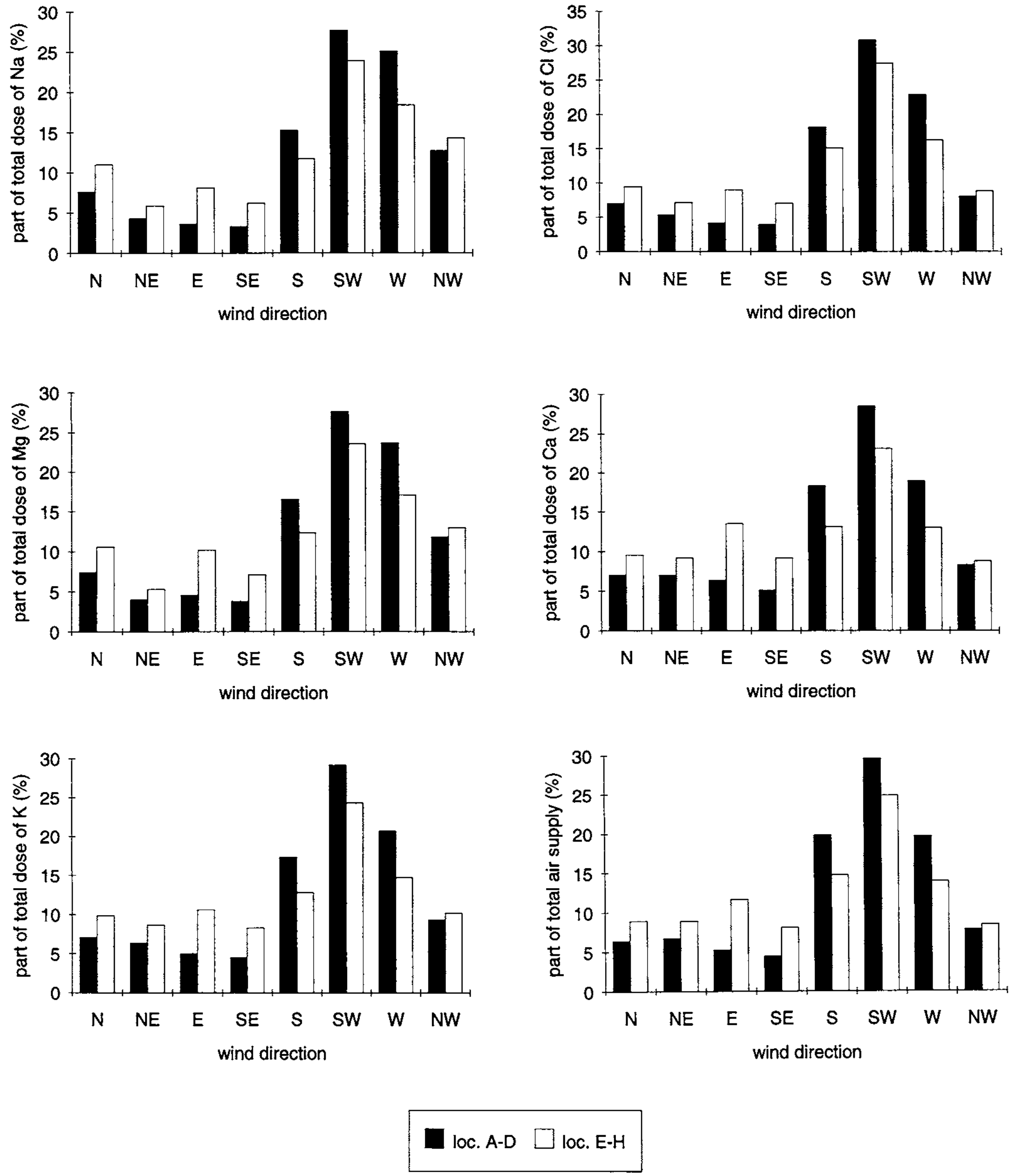


FIGURE 7.16 *(continued)* The percentage to which a particular wind direction contributed to the total pollutant dose of Na^+, Cl^-, Mg^{2+}, Ca^{2+} and K^+, and total air supply during the measurements, respectively

The computed *WEINTE* factors in relation to edge aspect are presented in Table 7.15. For SO_4^{2-}, NO_3^- and NH_4^+, the enhancement factors were found to range between 1.08 and 1.23. Edges with southeastern, southern, southwestern and western aspects showed the highest values. Enhancement factors for Na^+, Cl^- and Mg^{2+} were found to range between 1.23 and 1.66, where edges with southern, southwestern, western and northwestern aspects showed the highest values. Values presented in Table 7.15 rely on the assumption that representative mean enhancement factors were measured for forest edges with a southwestern aspect. However, mean structure characteristics of the eight forest edges (z_o = 1.5m, height = 13.9m and LAI = 6.2) may be considered more-or-less representative for an average Dutch forest. Moreover, the computation of *WEINTE* factors in relation to edge aspect was based on data (percentages to which particular wind directions contribute to the total pollutant dose) which most probably only hold for the central part of the Netherlands and at great distances from local sources. Finally, *WEINTE* factors presented only hold for linear forest edges. In case of non-linear edges, larger enhancement factors will occur as the disturbance of the wind profile will be larger. In the Netherlands linear forest edges are typical.

TABLE 7.15 Whole-edge integrated net throughfall enhancement (*WEINTE*) factors in relation to the edge aspect. x = mean of all aspects

	N	NE	E	SE	S	SW	W	NW	x
SO_4^{2-}	1.09	1.09	1.13	1.19	1.23	1.23	1.18	1.13	1.16
NO_3^-	1.08	1.08	1.10	1.14	1.18	1.18	1.16	1.11	1.13
NH_4^+	1.09	1.09	1.11	1.15	1.18	1.18	1.15	1.11	1.13
Na^+	1.38	1.27	1.25	1.36	1.52	1.64	1.66	1.54	1.45
Cl^-	1.33	1.25	1.28	1.42	1.59	1.66	1.64	1.49	1.46
Mg^{2+}	1.31	1.23	1.23	1.33	1.46	1.54	1.55	1.45	1.39
Ca^{2+}	1.15	1.13	1.14	1.19	1.24	1.25	1.24	1.19	1.19
K^+	1.05	1.04	1.04	1.06	1.08	1.09	1.09	1.07	1.07
Volume	1.02	1.01	1.01	1.02	1.03	1.03	1.03	1.02	1.02

The impact of edge effects on dry deposition amounts to forests in the Netherlands

A rough assessment of the percentage of Dutch forests influenced by edge effects was made with help of information available in the Dutch Forest Statistics (CBS, 1985). Two kinds of forest edges were distinguished, namely edges between forest complexes (defined as coherent sets of forest stands) and non-forested areas, and edges situated within forest complexes between forest stands of different heights. In Figure 7.17, the number and total area of forest complexes as well as individual forest stands in the Netherlands are presented by size class. In total, 23,871 forest complexes and 213,691 individual forest stands were present during the recording period 1980-1985. The total forested area in the Netherlands approximated 334,027 ha. Almost 80% of all forest complexes were found to be smaller than 5 ha; only 18 complexes were larger than 2000 ha. More than 70% of all forest stands were found to be smaller than 1.5 ha. These figures clearly indicate that Dutch forests are considerably fragmented.

Considering forest complexes as squares, their edge area was computed by:

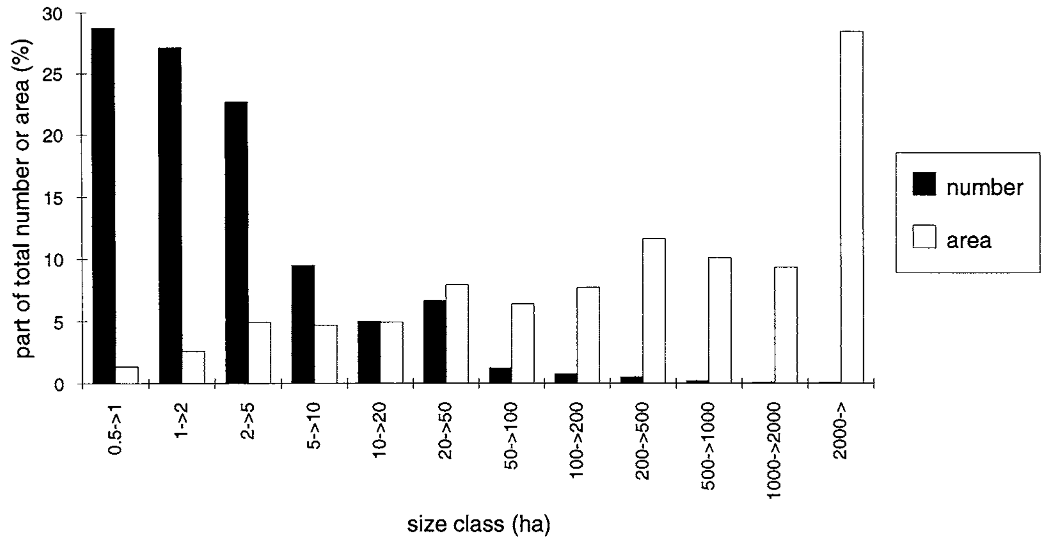
$$EA_{fc} = TA_{fc} - (TA_{fc}^{0.5} - 2L)^2 \qquad [7.3]$$

in which EA_{fc} is the edge area of the forest complex, TA_{fc} the total area of the forest complex, and L the edge width. With an average height of forest stands in the Netherlands of 11.7 m (Meijers, 1990), and an edge zone which approximate five edge heights, L equals, on average, 58.5 m. The percentage edge area [= (EA_{fc}/TA_{fc}) * 100%] of each size class was computed using the average TA_{fc} values for each size class. By weighting these percentages for the contribution of each size class to the total area of Dutch forests, it was calculated that 24% of the total forested area in the Netherlands consisted of forest edges bordering a non-forested area.

Similarly, the edge area between forest stands of different height was computed by:

$$EA_{fs} = 0.5 * (TA_{fs} - (TA_{fs}^{0.5} - 2L)^2) \qquad [7.4]$$

in which EA_{fs} and TA_{fs} are the edge area and the total area of the forest stand, respectively. A factor of 0.5 was added because it was assumed that a transition between two forest stands of different heights will cause enhanced dry deposition amounts in the highest stand only. From the frequency distribution of forest stand heights (Figure 7.18), it was computed that the average height difference between two forest stands equalled 8.0 m. A random distribution of forest stands was assumed. In reality, however, forest stands of the same age will be grouped together, resulting in a somewhat lower mean height difference. Because this effect could not be quantified, a mean height difference of 8.0 m was used for the computation of L (= 40 m). The percentage edge area [= (EA_{fs}/TA_{fs}) * 100%] of each size class was, similar to above, computed using average TA_{fs} values for each size class. By weighting these percentages for the contribution of each size class to the total area of Dutch forests, it was calculated that 28% of the total forested area in the Netherlands consisted of edges between two forest stands.



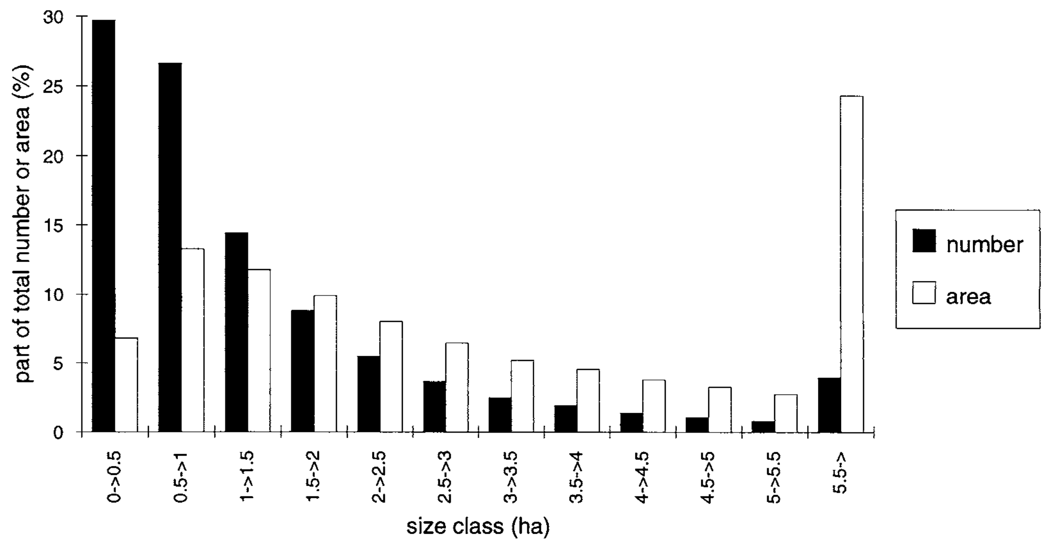


FIGURE 7.17 The percentage to which different size classes contribute to the total number (and area) of forest complexes (above), and to the total number (and area) of individual forest stands in the Netherlands (below).

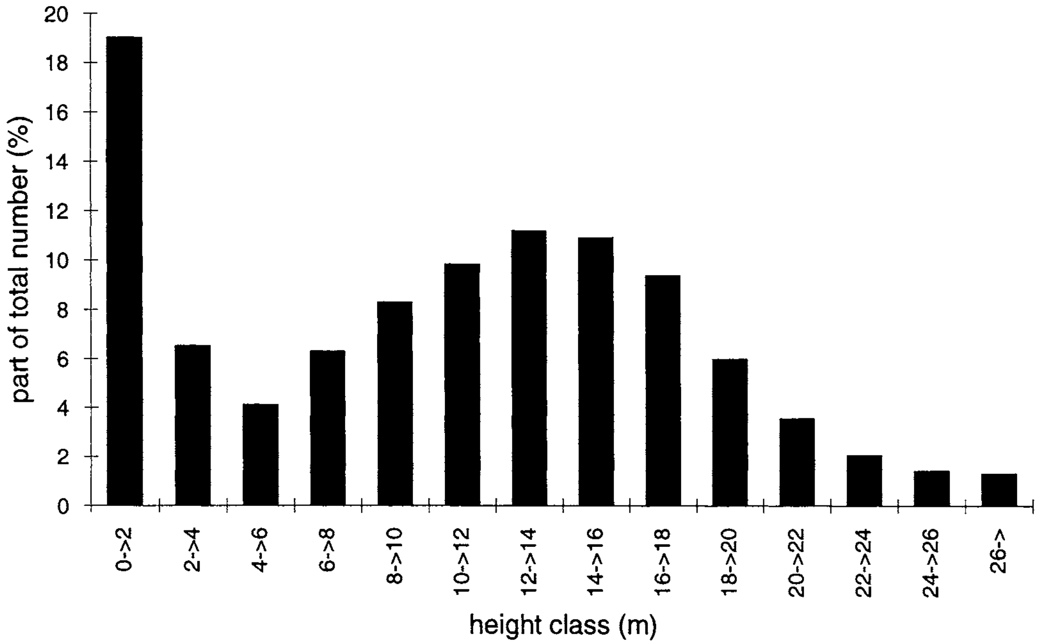


FIGURE 7.18 Frequency distribution of forest stand heights in the Netherlands. The relatively high percentage observed for height class 0-2 m is the result of the relatively high fraction of timber felling areas and non-afforested silvicultural areas in the Netherlands.

When an edge width of five edge heights is applied, the total percentage of Dutch forests influenced by edge effects thus amounts 52% (if other values for edge width are used the total percentage calculated will change according to Figure 7.19). This percentage is probably too low, because small forest roads, fire lanes and canopy gaps, also expected to cause edge effects, have not been considered in the calculations. Moreover, forests are considered as squares in the calculations, whereas in reality irregular-shaped forest complexes and rectangular-shaped forest stands also occur frequently. However, taking this 52% for granted, and assuming that *WEINTE* factors for SO_4^{2-}, NO_3^- and NH_4^+ presented in Table 7.15 are valid all over the Netherlands and representative for all kind of forest edges, it can be computed that by neglecting edge effects, dry deposition of acidifying compounds to forests in the Netherlands may be underestimated by approximately 5-10%. Similarly, it can be computed that dry deposition of sea salt particles may be underestimated by 20-25%. Most probably, these are upper limits of underestimation because enhanced dry deposition in a forest edge may lead to a significant downwind depletion of gases and particles, resulting in a relatively reduced deposition in the forest interior. Significant negative relationships found between *WEINTE* factors of Na^+, Cl^- and Mg^{2+} on the one hand, and their basic net throughfall fluxes on the other (R=-0.75, p<0.05; R=-0.72, p<0.05 and R=-0.75, p<0.05, respectively) may be an indication for this. Such relationships were not observed for SO_4^{2-}, NO_3^- and NH_4^+.

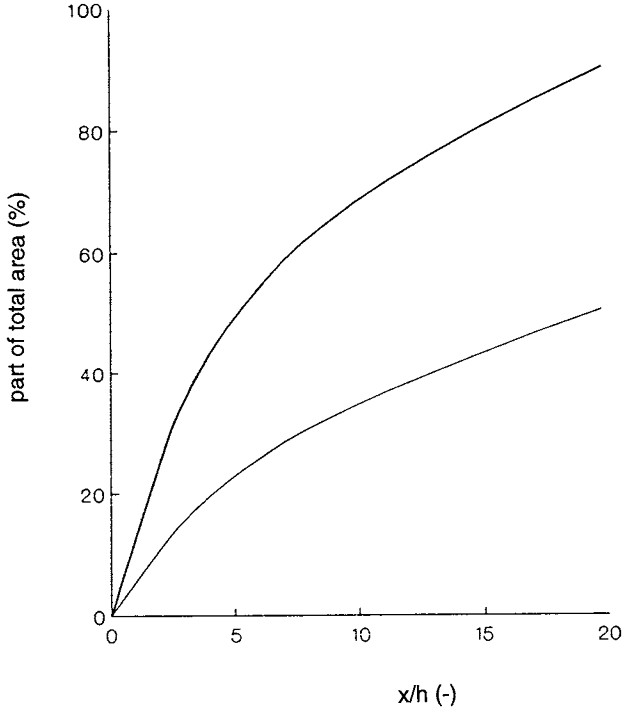


FIGURE 7.19 Relationship between the part of the total area of Dutch forests influenced by edge effects and edge width. The thin line represents this relationship when only edges between forested areas and non-forested areas are taken into account.

7.2.5 CONCLUSIONS

The impact of forest canopy structure on deposition amounts

Strong relationships exist between net throughfall fluxes of SO_4^{2-}, NO_3^-, NH_4^+, Na^+ and Cl^- on the one hand, and the roughness length and leaf area of the canopy on the other. For SO_4^{2-} and NH_4^+ this was explained by the relatively small surface resistance to SO_2 and NH_3 in the Netherlands, whereby dry deposition is controlled to a large extent by the aerodynamic roughness (length) of the canopy. For NO_3^- it was assumed that the surface resistance of NO_2 is inversely related to the leaf area of the forest stand, and/or that the net throughfall flux of NO_3^- was mainly determined by dry deposition of HNO_3 and NO_3^- aerosol. Dry deposition of these compounds depends largely upon atmospheric transfer to the receptor surface. Compared to acidifying compounds, net throughfall of Na^+ and Cl^- show weaker, but still significant, correlation with the roughness length and leaf area of the canopy. Net throughfall of these compounds is largely controlled by dry deposition of sea salt particles which, in turn, depends on, in addition to aerodynamic properties and the collecting surface area of the canopy, the canopy density and the collecting efficiencies of individual canopy elements. Relationships between net throughfall of SO_4^{2-}, NO_3^-, NH_4^+, Na^+ and Cl^- on the one hand, and *LAI* on the other, are linear in the *LAI* range of 2.4 to 12.0.

The impact of forest canopy structure on deposition amounts is very complex and must be separately considered for each component. Aerodynamic transport, quasi-laminar layer transport and surface resistance are influenced by canopy structure characteristics. Atmospheric transport from the free atmosphere to the forest canopy depends heavily on tree height and leaf area (determining the roughness length); transport through the quasi-laminar layer is influenced, for instance, by the leaf area density (controlling in-canopy wind speeds) and tree species (determining the collecting efficiency of individual canopy elements); the surface resistance is, among other factors, influenced by tree species and leaf area.

Incorporating detailed information on tree height, leaf area (density) and tree species in present-day inferential deposition models offers prospects for estimating deposition fluxes to individual forest stands more accurately. Such detailed information may be obtained through forest inventories or by using remote sensing techniques. Approximately every 15 years in the Netherlands a detailed forest inventory is made through which information on tree species, mean tree height and crown projection is available on stand level (CBS, 1985). In the near future this information will become available in a Geographical Information System (Stuurman, personal communication). Recent applications of remote sensing in air pollution research have focused on directly visible effects of atmospheric pollution, such as defoliation and yellowing of vegetation (e.g. Westman and Price, 1988; Vogelmann and Rock, 1986). However, reasonable relationships have also been found between reflection values of Landsat Thematic Mapper (TM) images and *LAI* (Peterson *et al.*, 1987). Tree species can also be detected from satellite data. Remote sensing seems to be a potentially useful tool for determining canopy structure characteristics from the viewpoint of deposition modelling (Heil *et al.*, 1992), especially when a combination of multi-temporal Landsat and Spot-images is used (Meijers *et al.*, 1993).

The impact of forest edge effects on deposition amounts

In general, net throughfall fluxes inside forests increase exponentially towards forest edges. The width of the zone with enhanced net throughfall fluxes approximates five edge heights. Net throughfall enhancement of SO_4^{2-}, NO_3^- and NH_4^+ near forest edges (on average, a factor of 2) is smaller than that of Na^+, Cl^- and Mg^{2+} (on average, a factor of 5). Differences are attributed to the sources for these ions in throughfall. Net throughfall of SO_4^{2-}, NO_3^- and NH_4^+ in the Netherlands is determined to a large extent by dry deposition of gases and submicron particles, and net throughfall of Na^+, Cl^- and Mg^{2+} largely by dry deposition of supermicron sea salt particles.

Net throughfall enhancement in forest edges also strongly depends upon forest density and edge aspect. For SO_4^{2-}, NO_3^- and NH_4^+, very strong relationships were observed between net throughfall enhancement factors and silhouette area density, suggesting that drag forces (influencing aerodynamic transport) mainly determine the differences between forest edge and forest interior dry deposition. For Na^+, Cl^- and Mg^{2+}, the largest correlation coefficients were found with leaf area density, which suggest that for these ions in-canopy wind speeds largely determine the differences between forest edge and forest interior dry deposition. Forest edges exposed to prevailing wind directions and/or high pollutant doses are especially subject to enhanced dry deposition. For SO_4^{2-}, NO_3^- and NH_4^+, the largest net throughfall enhancement factors were found in forest edges with SE, S, SW or W aspect. This was in agreement with results from an extensive survey of the groundwater quality under forests in the Netherlands (Bouwmans and Beltman, 1991). The largest SO_4^{2-} and NO_3^- concentrations in groundwater were found in forest edges exposed to SE, S, SW and W wind directions. For Na^+ and Cl^-, the largest net throughfall enhancement factors were observed in forest edges exposed to S, SW, W and NW wind directions.

In large parts of Europe, extensive uniform forests are more the exception than the rule and also Dutch forests are considerably fragmented. Almost 80% of all forest complexes in the Netherlands are smaller than 5 ha, and more than 70% of all individual forest stands smaller than 1.5 ha. For an edge width of five edge heights, it was computed that at least 50% of the total forested area in the Netherlands is influenced by edge effects. By neglecting edge effects, dry deposition of acidifying compounds to forests in the Netherlands as a whole may be underestimated by approximately 5-10%. Similarly, dry deposition of (sea salt) particles may be underestimated by 20-25%. These are upper limits of underestimation because it may be expected that enhanced dry deposition in a forest edge leads to a downwind depletion of gases and particles, resulting in relatively reduced dry deposition in the forest interior. Therefore it is desirable to obtain more insight on the impact of edge effects on regional deposition amounts. A combination of modelling and measuring surface fluxes in heterogeneous landscapes seems most promising for this purpose. Incorporating edge effects in present-day deposition models may offer prospects to estimate regional deposition amounts and deposition to individual forest stands more accurately. Most probably this will require the use of detailed geographical information on the length and exposition of existing edges, the height difference between edges, and the ratio between edge area and total area of forest stands (Meijers *et al.*, 1990). Such information should be available in a Geographical Information System (Burrough, 1986). Remote sensing techniques also seem useful for detecting forest edges and for quantifying landscape heterogeneity (Heil *et al.*, 1992).

7.3 THE SPEULDER FOREST EXPERIMENTS TO DETERMINE THE INPUT AND RELATED IMPACTS TO DOUGLAS FIR

7.3.1 INTRODUCTION

During the past nine years (1986-1994) research has been conducted at the Speulder forest at the Hoge Veluwe in the centre of the Netherlands. Most research was part of the Dutch National Programme on Acidification, which has now reported its third and probably final phase (Heij and Schneider, 1995). The results of the first two phases were summarised in Schneider and Bresser (1988) and Heij and Schneider (1991). The main emphasis of the research at the Speulder forest was on acidification. The research and monitoring programmes aimed: *i)* to estimate current loads and levels of air pollutants such as sulphur and nitrogen compounds, but also base cations, *ii)* to determine forest characteristics and follow growth parameters in time, *iii)* to determine the effects or risks in relation to exposure of high pollutant loads and levels and *iv)* to determine the effects of reduction in these loads and levels.

Deposition research

In 1990 a large project was initiated to develop and evaluate a monitoring method for measurement of deposition of acidifying components onto forests. This project represents the continuation of the successful development of monitoring methods for SO_2, NO_2 and NH_3 deposition to low vegetation (Erisman, 1992; Mennen *et al.*, 1992; Erisman *et al.*, 1993a, 1993b; Wyers *et al.*, 1993a; Erisman and Wyers, 1993; see also Section 7.1). The Speulder forest was chosen because most of the infrastructure was already present after the site had been used for a three-year experiment. Dry deposition estimates were derived from these measurements by Vermetten *et al.* (1992). However, several problems were encountered in the analysis of the data because the accuracy of the measured gradients was too small. This was the consequence of an experimental set-up which was not aimed at accurate measurement of deposition. Duyzer (1992) made a systematic analysis of possible errors and recommended improvements for dry deposition monitoring onto forests.

From January to August 1992 tests were done over low vegetation on the heathland, Elspeetsche Veld (Zwart *et al.*, 1994) with the monitoring equipment. The purpose was *i)* to determine effects of obstacles (monitor housing) to be installed in the mast above the forest on the momentum and heat flux measurements, *ii)* optimisation of the gradient system for NO_2 and SO_2, *iii)* comparison of SO_2 dry deposition parameters with those measured with another dry deposition monitoring system (Mennen *et al.*, 1995), and *iv)* tests of eddy correlation measurements of the NO_2 flux. At the end of 1992 the optimised monitoring systems for SO_2, NO_2 (RIVM) and NH_3 (ECN) were installed at the forest site. Since then continuous vertical concentration gradient measurements of these components have been available. In June 1992 eddy correlation measurements of NO_2 were also started. From that time on, direct NO_2 flux measurements have been available. These data, however, have not yet been validated and evaluated.

Next to gaseous deposition measurements, a project was run for six months to determine the particle flux at Speulder forest and to compare atmospheric deposition with throughfall measurements (Erisman *et al.*, 1994). The three main research issues addressed in this project were:
- What is the contribution of 'acidifying' aerosols to the total acid input of nature areas?
- What is the relation between atmospheric deposition estimates and throughfall measurements and what is the contribution of aerosol deposition to the difference between the two estimates?
- How important is the coarse particle flux (base cations) to the nutrient cycle in nature conservation areas?

The research required for satisfying answers to the three research issues was defined by a project group, in which research institutes and universities with experience in the field of aerosol research in the Netherlands participated. The work on the 'aerosol project' was a joint initiative of the *National Institute of Public Health and Environmental Protection (RIVM), KEMA (Laboratory for Environmental Research), TNO (Institute of Environmental Sciences), ECN (Netherlands Energy Research Foundation), RUU-FG (Utrecht University, Department of Physical Geography)* and *WAU-AQ (Wageningen Agricultural University, Department of Air Quality)*. First, a model was selected from existing models of aerosol deposition to forests (Ruijgrok *et al.*, 1994). The model was to be representative for the Dutch situation (pollution climate). Insight from model simulations into the most important processes involved in aerosol deposition was gained. The main processes were tested by means of experimental research in the field (at the Speulder forest). The results of the experiments led to a verification of the model description and a basis for a parametrisation of V_d in terms of routinely available data. The parametrisation was used for the generalisation of aerosol deposition to other nature conservation areas in the Netherlands. It was anticipated in the project that by executing all available experimental techniques in combination with a large modelling effort, a more accurate estimation of particle dry deposition velocities for rough surfaces would be obtained. In June 1993, TNO organised an international field campaign in the Speulder forest. The University of Manchester (UMIST) and RISOE National Laboratory (Denmark) participated in a three-week campaign. During the campaign particle fluxes were measured by UMIST using an eddy correlation method. The preliminary results have been kindly made available for use.

Assessment of relations between loads/levels and effects

The main focus of this section will be on the determination of atmospheric input and soil loads at Speulder forest. However, at the end, the major findings of the research at the Speulder forest will be used to assess the causal relations between loads and levels on the one hand and effects on the other. First, the atmospheric loads and levels at the Speulder forest will be presented. The uncertainty in the atmospheric load and level estimates will be discussed along with their evolution during the past years. Subsequently, critical loads and levels derived for the Speulder forest and exceedances determined will be outlined. Critical levels and loads refer to thresholds, which can serve as a tool in assessing the occurrence of effects in natural ecosystems (Nilsson and Grennfelt, 1986; Hettelingh *et al.*, 1991; see Chapter 1 for definitions).

Finally, effects observed at the Speulder forest will be discussed and the concept of critical loads and critical levels evaluated. Effects are defined as ecosystem changes due to environmental impacts. Risks for adverse effects in the future are indicated. Results from manipulation experiments are used to show the recovery of forests after reducing soil loads to pre-industrial levels. At the end of this section, the research programme carried out at the Speulder forest since 1985 will be critically evaluated on its merits and shortcomings.

7.3.2 SITE DESCRIPTION

The Speulder forest is located in the Veluwe, a large undulating area with forests and heathlands in the central part of the Netherlands. The measuring site covers an area of 2.5 ha, planted with 2 year old Douglas fir trees (*Pseudotsuga menziessi* (Mirb.) Franco L.) of the provenance Arlington in 1962. Gaps caused by windthrow and diseases were filled with Douglas fir of unknown provenance. The canopy is well closed, with the exception of some gaps due to windthrow caused by the heavy storms in February 1991. The one-sided LAI was between 13.9 and 9.7 for the measuring years 1987-1993 (Steingröver and Jans, 1995). The stand is surrounded by a forested area of approximately 50 km^2. The stand itself is surrounded by Larix, birch, Pedunculate oak, Scotch pine and Douglas fir stands, with mean tree heights varying between 12 and 25 m. A small clearing of 1 ha is situated to the north of the stand.

In 1986, when the stand was selected, the mean number of needle year classes on an average first-order branch in the sun-adapted crown level of a tree was four. According to European Community vitality parameters, the vitality of the stand was better than the nationwide average for Douglas fir (Steingröver and Jans, 1995). The stem density varies between 765 trees ha^{-1} in the eastern part of the stand and 812 trees ha^{-1} in the western part. Unless stated otherwise, all measurements took place in the eastern part of the stand. At the end of 1993 the average *DBH* was 25 cm and the trees were approximately 22 m height (Jans *et al.*, 1994).

Soil chemical and texture data are given in Table 7.16. The forest stand is situated on top of an ice-pushed ridge. The groundwater is found at a depth of about 40 m. The soils are well drained. The soil is a Typic Dystochrept on sandy loam and loamy sand-textured Rhine sediments of Middle Pleistocene age. The soil can also be classified as an Orthic Podzol (FAO, 1988) or as a Holtpodzol (Van Breemen and Verstraten, 1991). The soil is rather heterogeneous. The texture of the soil shows a strong spatial variability related to the elongated, parallel outcrops of layers of different textures typical of an ice-pushed ridge.

TABLE 7.16 Soil chemical and texture data for the Speulder forest (Van Breemen and Verstraten, 1991)

Depth (cm)	pH H_2O	pH KCl	%C	%N	*CEC*[a] mmol/kg	Texture (%) <2 μm	2-16 μm	16-2000 μm	> 2000μm
0 - 5	3.63	2.83	7.3	0.30	59	1	3	70	1
5 - 10	3.70	3.00	2.9	0.11	39	0	3	87	0
15 - 20	3.87	3.70	2.0	0.07	42	0	4	94	1
30 - 35	4.15	4.27	0.8	0.04	21	0	3	95	2
50 - 55	4.22	4.38	0.3	0.02	13	0	2	97	0
90 - 95	4.22	4.28	0.2	0.01	24	0	2	95	0

[a] *CEC*: calculated as the sum of 0.5 M $BaCl_2$ extractable Ca^{2+}, Mg^{2+}, K^+, Na^+, plus 1 M KCl extractable Al^{3+}, H^+ and NH_4^+.

The climate is moderately humid with an average precipitation of around 800 mm a^{-1} Large sources of SO_2 and NOx are located 200 km to the southeast (industrial Ruhr area) and 100 km to the southwest (Rotterdam port). The distance to NH_3 sources varies from a few kilometres to the south to some 10 kilometres to the north of the stand.

7.3.3 RESEARCH PROJECTS

Several investigations were conducted at the Speulder forest. Until 1990, these were co-ordinated by ACIFORN, established to analyse and quantify the effects of air pollution and soil acidification on forest growth and vitality (Evers *et al.*, 1991). The results of the projects conducted within the framework of ACIFORN have been reported in Heij and Schneider (1991) and in Evers *et al.* (1991). After 1990 research at Speulder forest was not co-ordinated, but the location was extensively used for different kinds of investigations. One of the research topics since 1990 has been the investigation of deposition of acidifying pollutants and base cations. The research activities at the Speulder forest site related to deposition research are summarised in Figure 7.20 (section 7.3.4).

At the Speulder forest research was also conducted on: *i)* boundary-layer clouds and vegetation-atmosphere exchange (KNMI), *ii)* the physiology of Douglas fir trees (IBN-DLO), *iii)* the effects of manipulation of nutrient inputs, (see Figure 7.20, KUN-NITREX), *iv)* mono-terpene emissions from trees, chlorine formation in the soil and chloroform emission from the soil (TNO) and *v)* the hydrological cycle in the forest and the forest soil (UvA FGBL). The primary aim of the physiology project was to analyse and quantify the physiological effects of air pollution, drought and nutrient supply/availability on trees under field conditions (Steingröver and Jans, 1995). The project descriptions of these studies can be found in Heij and Schneider (1992).

7.3.4 DEPOSITION MONITORING OF GASEOUS COMPONENTS

Introduction

Erisman *et al.* (1993) evaluated the deposition measurements made between November 1992 and September 1993. The main emphasis was on the evaluation of the dry deposition measurements for routine application, estimation of fluxes and their accuracy, and deriving a parametrisation for the surface resistance to deposition on forest. Dry deposition parameters were derived from the results of the hourly average measurements. Following the recommendations reported by Erisman *et al.* (1993), the system configuration and the measurement scheme were adjusted. The system configuration, several tests, and the calibration and quality control procedures are extensively described in Mennen *et al.* (1995). In this section, a summary of the evaluation of the deposition monitoring methods and the interpretation of data will be described along with the results of two years of monitoring.

Experimental procedure

The Speuld location was equipped with two towers and measuring facilities. One tower was used for gas deposition measurements. The second tower was used for the so-called 'Aerosol project' (Erisman *et al.*, 1994; see section 7.2.5). A schematic representation of the monitoring system at the Speulder forest is shown in Figure 7.20. Most of the equipment is put on a tower consisting of a mast, a scaffolding and two hoists, both provided with a suspension frame. One of the hoists is used for transport of material, whereas the other is used to carry two boxes containing the monitors, which can be transported downwards for calibration or maintenance. Both mast and scaffolding are 36 m high and have a triangular cross-section (Zwart *et al.*, 1993; Mennen *et al.*, 1995).

A sonic anemometer (Kaijo Denki DAT-310) mounted on the top of the scaffolding at 36.5 m height was used to measure the horizontal and vertical wind velocity, wind direction and friction velocity, and the sensible heat flux. A net radiation meter (Thies 8110) and a temperature and relative humidity sensor (Vaisala HMP121B) were mounted 1.5 m outside the scaffolding towards the south, at heights of 35 and 33 m, respectively. The two boxes housed the gas monitors, two pulsed fluorescence SO_2 analysers (Thermo Environmental Instruments Inc., Model 43S) and two Luminox NO_2 analysers (Scintrex Ltd., LMA-3). The performance of the instruments and the monitoring system as a whole was thoroughly tested by Zwart *et al.* (1993). This work also contains an extensive description of the system. Zwart *et al.* concluded that the performance of the SO_2 monitors is sufficient for dry deposition monitoring. For NO_2 they demonstrated that the performance to be questionable; it should be further tested in the field.

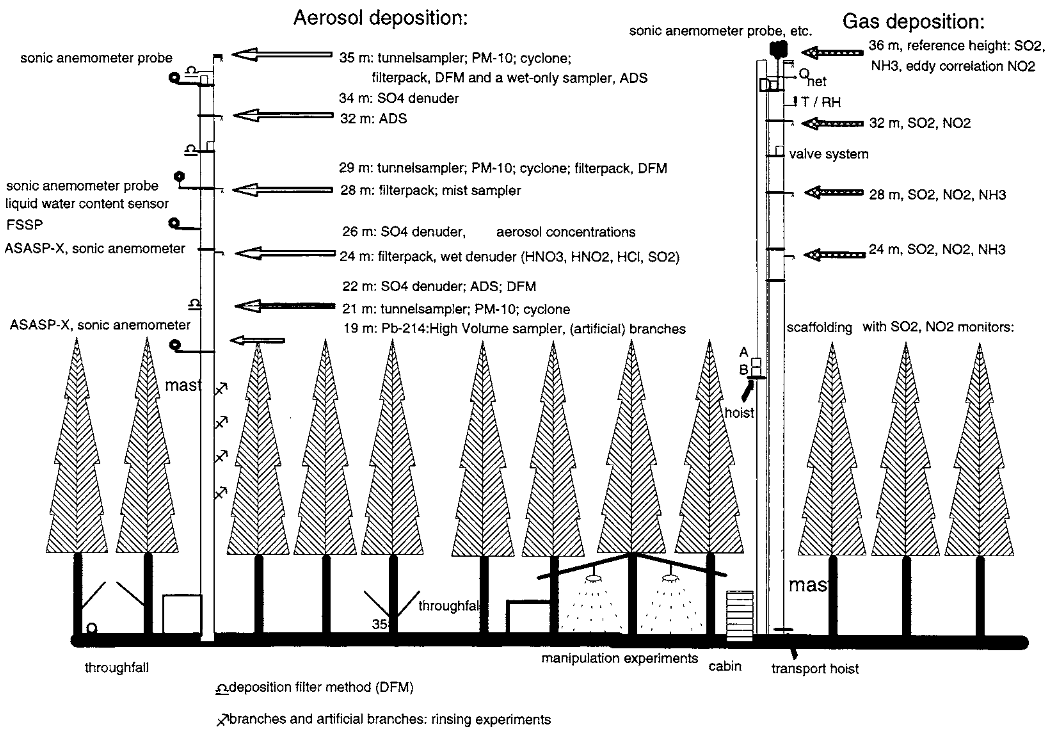


FIGURE 7.20 Deposition research at the Speulder forest site.

Theory and interpretation

Dry deposition of gases can be calculated from concentration gradients and meteorological parameters. The calculation of c_*, F , V_d and Rc is outlined in Chapter 3. c_* is obtained from the weighted regression between c and the stability corrected height (4 levels). Weights are obtained from the standard deviation of the 1-min concentration measurement at each height. The concentrations of SO_2 and NO_2 are corrected by variations in time using the concentrations measured with the reference monitor (M_r) by:

$$c(M_s, t_1, z) = c(M_s, t_i, z)\frac{c(M_r, t_1, 36)}{c(M_r, t_i, 36)} \qquad [7.5]$$

t_i denotes the successive four measuring periods for determination of the gradient by the scanning monitor (M_s).

During the experiments in 1989 at the Speulder forest, the Royal Netherlands Meteorological Institute (KNMI) compared gradients of temperature and wind speed above the forest with directly measured fluxes of heat and momentum in order to test the commonly used flux profile relations (Dyer and Hicks, 1970). From these experiments systematic differences were found between the Dyer and Hicks relations and those obtained above the Speulder forest (Bosveld, 1991; Duyzer *et al.,* 1992). This can be accounted for by introducing α_h, a height-dependent correction factor. In the roughness layer the following relations were proposed for the flux profile relations for heat (Bosveld, 1991):

$$\phi_h = \alpha_h \, \phi_h, \text{ for unstable conditions}$$
$$\phi_h = \phi_h - (1-\alpha_h), \text{ for stable conditions} \qquad [7.6]$$

TABLE 7.17 Correction factor for flux profile functions over the Speulder forest (Bosveld, 1991).

Height intervals (m)	α_h
18-24	0.65
24-31	0.80
31-36	0.95

α_h is a function of height and equals unity at the height where the influence of the individual roughness elements inducing extra turbulence has become small (for the Speulder forest this is at z≈36 m). The correction factors α_h for the flux profile function for heat are given in Table 7.17.

It is assumed here that these factors can also be applied for trace gas flux profile relations. The method of application proposed by Bosveld (1991) and Duyzer *et al.* (1992) is adopted here. Flux profile relations for ozone and heat were evaluated at the Speulder forest by TNO. They showed that similar alpha factors can be applied for ozone (Westrate and Duyzer, 1994).

Dry deposition parameters for the Speulder forest

Measurements of SO_2, NO_2 and NH_3 and meteorological parameters have been recorded ever since the end of November 1992. Because of problems with the NO_2 monitors, very little data are available to evaluate the gradients and to obtain fluxes. The main focus in this section is therefore on SO_2 and NH_3.

Constant flux layer assumption

The eddy correlation measurements of $u*$ and H at 30 m height by TNO and those at 36 m height by RIVM were compared to evaluate the constant flux layer assumption. Figures 7.21 and 7.22 show scatter plots of hourly average $u*$ and H values measured at the two heights in

December - June. A selection is made because during several weeks in December, February and March, the eddy correlation measurements by TNO were disturbed by an interfering signal on the datalogging system. Another selection was made for measurements with wind coming from the north through the clearing in the forest. Finally, u_* values below 0.1 m $^{-1}$ were not taken into account. About 50% of the dataset remains after selection. There is a reasonable agreement between the 30-m and 36-m u_* values; most scatter is due to local influence on the fetch and to scatter in the data as a result of differences in averaging time. The difference between the heat fluxes measured at the two levels was much larger. The reason for this large difference is that during and after rain or fog, heat flux measurements might be contaminated as a result of droplets on the sensors. Furthermore, at very low heat flux values (very stable conditions) the change on temperature inversions and thus decoupling of the air in and above the forest increases. Therefore, hours with differences larger than 25 W m^{-2} between the heat fluxes measured at the two heights were not taken into account. About 40% of the data remain; see plotted in Figure 7.22. From the two figures it is obvious that during these months the constant flux layer assumption for momentum and sensible heat fluxes is valid.

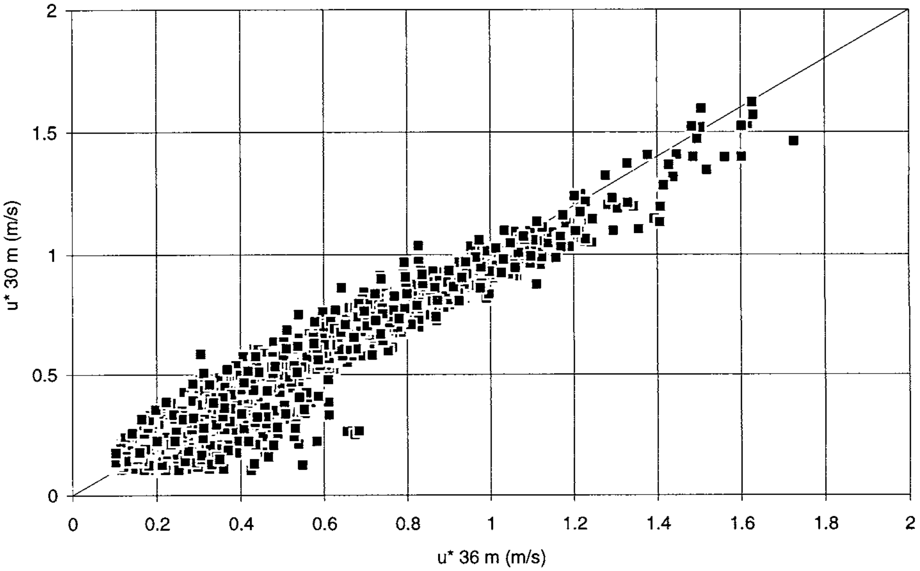


FIGURE 7.21 Eddy correlation u_* measurements at 30 m compared to those measured at 36 m.

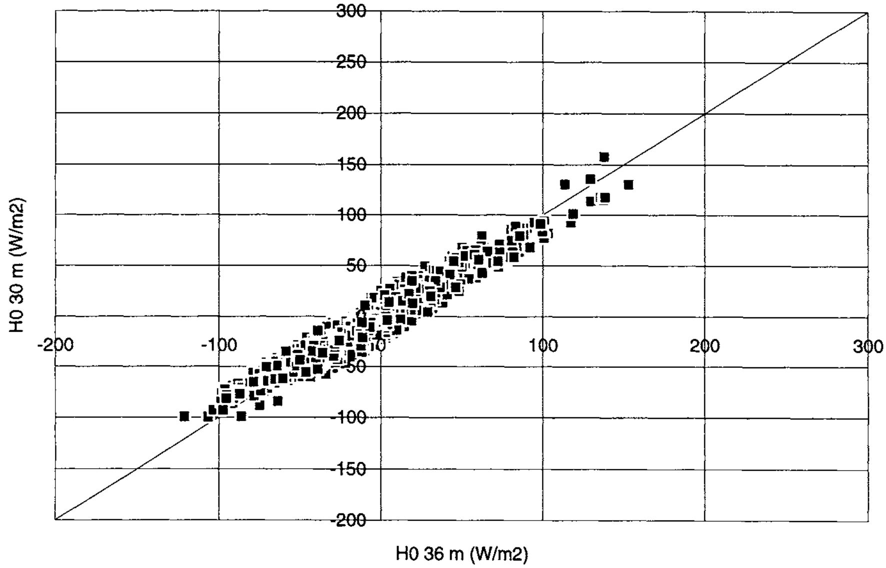


FIGURE 7.22 Eddy correlation H measurements at 30 m compared to those measured at 36 m.

Averages for selected periods

The three vertical SO_2 and NO_2 gradients were averaged for each hour and $c*$, F, V_d, R_a, R_b and R_c were calculated. The 10 NH_3 gradients measured within one hour were also averaged and the deposition values were calculated accordingly. The dataset thus obtained has to be 'cleaned' by selecting the hours during which: 1) the theoretical demands for the gradient technique were fulfilled; 2) the concentrations were well above the detection limit, and 3) there was no loss of necessary measurements due to technical problems. Selection criteria for SO_2 derived from measurements over a heathland (Erisman *et al.*, 1993a,b) were adjusted and applied here. These criteria are listed in Table 7.18. In this table the per cent of the total number of hourly averaged measurements remaining after applying the selection criteria is also given. For SO_2 about 42% survived the selection criteria. Most of the data were rejected as a result of measuring errors due to moisture in one of the sampling tubes (36-m height) and due to the random error in individually measured concentrations. It was found that by loss of one of the filters during a storm, the inside of the tubes had become moist, leading to adsorption and desorption of SO_2. This problem has now been overcome by heating the tubes. At concentrations below 5 $\mu g\ m^{-3}$ the random error is the dominating error in deposition parameters.

TABLE 7.18 Selection criteria for gradients measured over the Speulder forest and the percentage of measurements left after selection (total remaining: 2345 hours of continuous SO_2 measurements, 220 hours of NO_2 measurements and 756 hours of continuous NH_3)

Criteria	Percentage of remaining SO_2 data	Percentage of remaining NO_2 data	Percentage of remaining NH_3 data
Validity of flux profile relations u>1or u_*>0.1 m/s, plus NH_3 instrumental failure due to freezing	95	95	30
Fetch requirements: θ<330 and θ> 30°	89	89	
Deviation between two monitors: $\lvert\Delta C\rvert < 3\ \mu g\ m^{-3}$	78	21	-
No gradient: $c_* > 0$	77	-	-
Moisture in the tubes: 5-25 January 1993	59	-	-
High concentration fluctuations in time: dC/dt	58	-	28
High u* fluctuations in time: du_*/dt; σ_{u*}	57	-	-
Detection limit and random errors: $C > 5\ \mu g\ m^{-3}$	48	-	27
Stability range (errors in R_a and R_b): $\lvert[z-d]/L\rvert<0.5$	42	-	26

Most NO_2 data (80%) were rejected as a result of instrumental failure. It is obvious from this large amount of data lost that monitoring of NO_2 deposition with the current system is impossible. The Scintrex monitors are not suitable for routine application. In the meantime eddy correlation measurements with the Scintrex monitors have been started. Evaluation of these measurements is currently being carried out.

For NH_3, data from the period 28 November 1992 - 31 March 1993 have so far been processed. About 26% of the data remained after selection, see Table 7.18. Most data were rejected as a result of incorrect operation of instrumentation due to freezing of the solutes in the denuders. Application of this criteria and of the last two selection criteria in Table 7.18 may have lead to a bias in the dataset towards larger deposition velocities.

SO_2

Figure 7.23 shows the average daily variation in R_c values for selected hours for wet and dry leaf surfaces. During dry conditions a weak daily variation can be seen following stomatal behaviour. It must be emphasised that the standard deviations in the values presented in Figure 7.23 are very large (see Table 7.19). This has a large influence on the variation shown in Figure 7.24. During wet conditions R_c values are generally low. The average V_d and R_c values for dry and wet conditions, and for daytime and night-time are listed in Table 7.19.

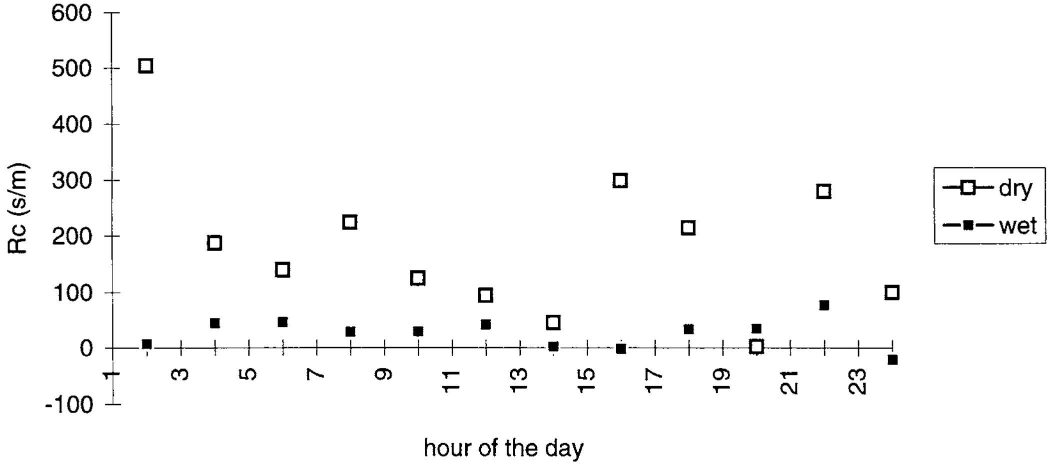


FIGURE 7.23 Daily variations of SO_2 in R_c for dry and wet surface conditions.

TABLE 7.19 Average deposition parameters for SO_2, with standard deviations in parentheses

	R_c (s m^{-1})		V_d (m s^{-1})	
Conditions	Observations	Modelled	Observations	Modelled
Night, dry canopy	250 (900)	90 (60)	0.008 (0.012)	0.012 (0.009)
Day, dry canopy	290 (2520)	150 (320)	0.007 (0.018)	0.011 (0.006)
Night, wet canopy	20 (120)	5 (10)	0.024 (0.021)	0.030 (0.017)
Day, wet canopy	20 (80)	5 (10)	0.030 (0.020)	0.027 (0.014)

Surface resistance parametrisation for SO_2 An R_c parametrisation derived from analogous measurements over a heathland during a three-year period (Erisman *et al.*, 1993a,b; section 7.1 and Chapter 4) was tested. This parametrisation is evaluated in section 6.1. Figure 7.24 shows 'measured' and parameterised SO_2 R_c values, and measured NH_3 R_c values for four successive days. Also shown in this figure are the concentrations of the two gases, the relative humidity and information on surface wetness. Parameterised and 'measured' R_c values for SO_2 show reasonable agreement, with high values during dry periods at night, low values during daytime and values approaching zero during wet surface conditions. R_c values for NH_3 show similar variations. For both this period and the whole dataset no influence of NH_3 on deposition parameters for SO_2 and vice versa was observed. This is in contrast to the observations of Erisman and Wyers (1993) who demonstrated influence of both gases on each other's deposition under extreme conditions. The explanation is that during November 1992 to April 1993 these extreme conditions did not occur. NH_3 is therefore not included in the parametrisation. For the dataset with the selected measuring periods, 40% of the variance in parameterised V_d versus the 'measured' V_d was accounted for with no systematic differences. These results are similar to those found for heathland vegetation using the same parametrisation (Erisman *et al.*, 1993a,b). A plot of the parameterised and measured V_d is given in Figure 6.1.

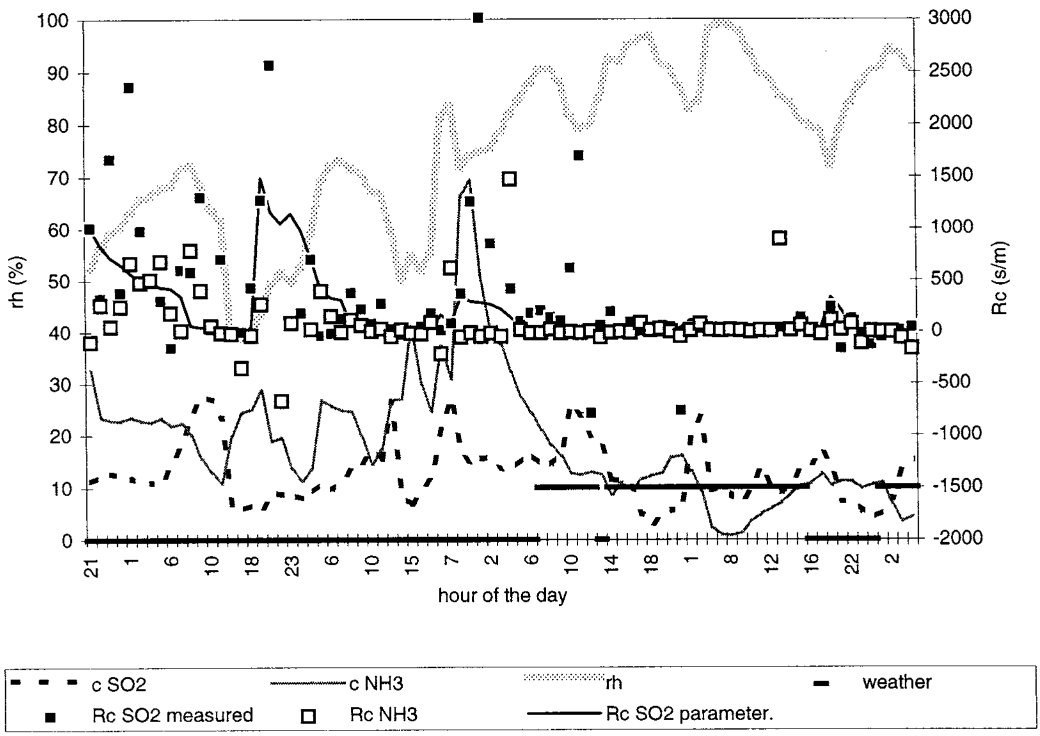


FIGURE 7.24 R_c values for SO_2 and NH_3 from continuous measurements of vertical gradients over the Speulder forest ('Weather' provides information on precipitation: 0 = dry; 10 = rain).

The surface resistance parametrisation will be used to estimate deposition parameters for measurements which did not satisfy the selection criteria. In this way, it is possible to estimate annual average deposition parameters using both the selected and the rejected data. One condition, however, is that this selection does not lead to a systematic bias in the parametrisation as a result of omitting an important deposition regulating process. When one year of monitoring data become available, this will be investigated by comparing surface conditions and measured parameters in selected and rejected periods, and looking for systematic differences (Erisman *et al.,* 1993c).

NO_2

Very few NO_2 measurements satisfy the selection criteria (Table 7.18). Most of the data has been rejected due to instrumental failure. The Scintrex NO_2 monitors are not suitable for routine applications. Currently, this is the only instrument which can measure NO_2 concentration with enough accuracy and speed for eddy correlation and gradient measurements (Zwart *et al.*, 1993). In 1994 results of laboratory tests with two new instruments which might be suitable would become available.

This section presents the first results of an analysis of the selected measuring periods. Figure 7.25 shows the average diurnal variation of V_d derived from the NO_2 concentration measurements satisfying the selection criteria. Also plotted in this figure is an estimate of the average diurnal variation of the deposition velocity as a result of stomatal uptake. The latter is calculated using Eqn. (4.4). Flux measurements over low vegetation and forests have shown that NO_2 dry deposition is determined by stomatal uptake (Hicks *et al.*, 1989; Duyzer 1992; Hargreaves *et al.,* 1992). A positive correlation between the V_d for stomatal uptake and the dry deposition velocity would therefore be expected. In Figure 7.25, however, an anti-correlation is observed with negative 'measured' values during daytime and positive (deposition) values at night, in contrast to positive stomatal values during daytime and zero values at night (stomata closed). The anti-correlation might be the result of NO emissions out of the forest, reacting with O_3 during daytime, forming NO_2 and thus establishing a net upward flux (Duyzer, 1992).

NH_3

The results of the NH_3 measurements have been reported in Wyers *et al.* (1993b). It is found that the surface resistance to ammonia deposition disappears when the canopy becomes wet (e.g. Figure 7.24). Emission of NH_3 was observed on several occasions and seemed to be strongly related to drying of the canopy. For the four-month period considered here (wintertime), it is estimated that as much as 20% of the deposited NH_3 was re-emitted from the canopy. Emission was observed mainly during the day at a relative humidity decreasing below 80%. So far, no relation is apparent between occurrence of emission and low or sharply decreasing ambient NH_3 concentrations. When the canopy is dry and the flux is towards the surface, this flux is higher than the inferred stomatal flux (Eqn. 4.4), indicating that the external leaf surface is also an important receptor for NH_3.

When the canopy is continuously wet, the surface resistance derived from the measurements is consistently negative with a typical value of -20 m s^{-1}. This indicates that the derived dry deposition velocities exceed the maximum possible V_d given by: $(R_a+R_b)^{-1}$. It is possible that R_a and/or R_b are overestimated under these conditions.

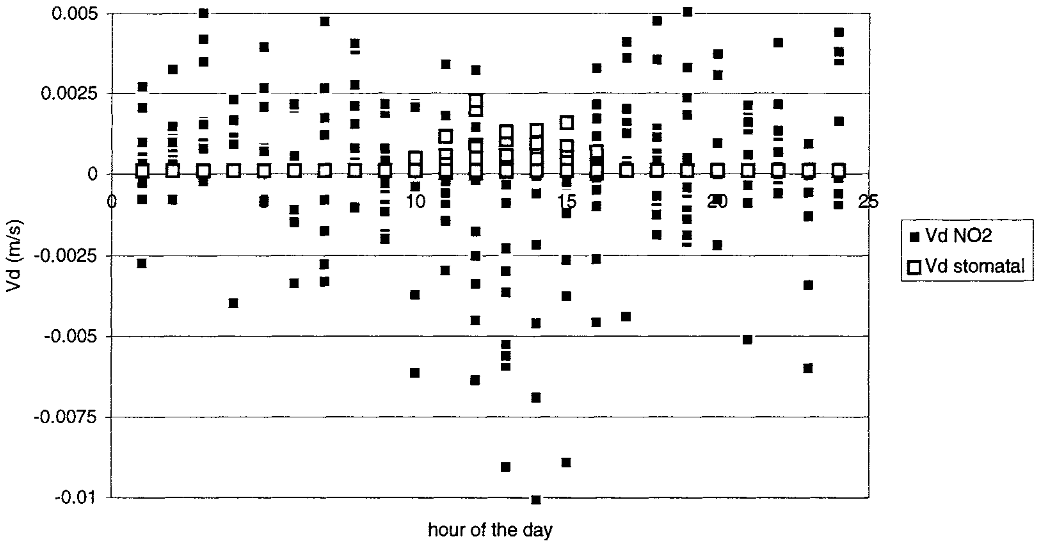


FIGURE 7.25 Diurnal variation of V_d NO_2 and V_d for stomatal uptake for selected measuring periods.

Measurements made at this site by Bosveld (personnel communication) suggest that R_b for water vapour is negligibly small. This might also be the case for NH_3 as its behaviour is similar. Another possibility is the presence of an additional sink above the canopy caused, for example, by chemical reactions or deposition to acid particles (Erisman and Wyers, 1993). Erisman and Wyers (1993) estimated that when using simulated water vapour, temperature, nitric acid and NH_4NO_3 aerosol gradients over heathland, negative R_c values could be the result of a shift in the equilibrium between gas phase and aerosol concentration between one height and the other above the surface.

Surface resistance parametrisation A start was made with the derivation of a parameterised description for NH_3 surface exchange (Wyers *et al.,* 1993b). Under conditions of deposition it was assumed that R_c could be described by:

$$R_c = \frac{1}{\frac{1}{R_{stom}} + \frac{1}{R_{ext}}} \qquad [7.7]$$

in which the stomatal resistance is described with Eqn. (4.4) and R_{ext} is the resistance to deposition for external leaf surfaces. For a wet canopy the R_{ext} is zero; for a dry canopy, R_{ext} is empirically derived as 20 s m^{-1}. The deposition velocity is calculated using Eqn. (3.3).

For a description of periods with NH_3 emission, related to evaporation of water films on the canopy, it is necessary to have an accurate description of canopy wetness and canopy drying. Unfortunately, this is not yet available. For the moment, emission is simply assumed to occur at global radiation above 0 W m^{-2} (daytime) and at a relative humidity below 80% during dry weather conditions. The resistance concept is not applicable for emission fluxes. However, as a first approximation, the 'emission' velocity is simply estimated using Eqn. (3.3) in a revised form:

$$V_e = \frac{-1}{R_a + R_b + R_{c,e}} \qquad [7.8]$$

with $R_{c,e}$ empirically derived as 10 s m^{-1}. Figure 7.26 shows the comparison between measured and modelled V_d. The prediction of occurrence of emission is still inadequate, as expected from the poor parametrisation for drying the canopy. Under conditions of deposition there is reasonable agreement between modelled and 'measured' V_d values, with 25% of variance accounted for.

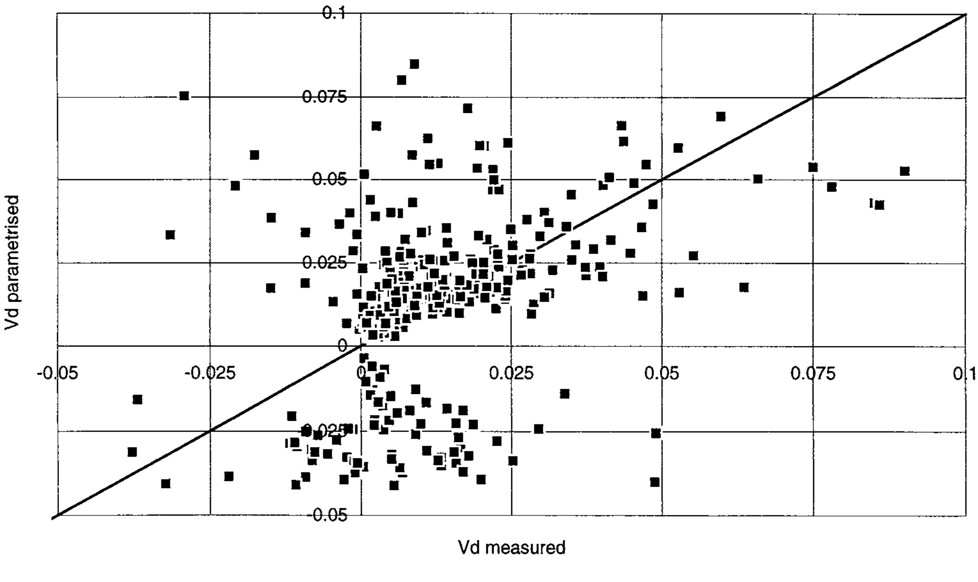


FIGURE 7.26 Parameterised values of V_d compared to V_d derived from NH_3 gradients for selected measuring periods (R^2=0.25 for positive values)

TABLE 7.20 Average deposition parameters for NH_3, with standard deviations in parentheses

	R_c (s m^{-1})		V_d (m s^{-1})	
Conditions	Observations	Modelled	Observations	Modelled
Night, dry canopy	20 (100)	20 (0)	0.02 (0.03)	0.02 (0.01)
Day, dry canopy	10 (40)	10 (0)	0.01 (0.05)	-0.01 (0.03)
Night, wet canopy	0 (30)	0 (0)	0.04 (0.05)	0.04 (0.02)
Day, wet canopy	0 (40)	0 (0)	0.04 (0.06)	0.04 (0.04)

In Table 7.20 observed and modelled averages of V_d and R_c are listed for the entire dataset. In general, there is good agreement between averages of observed and modelled values of the two parameters. An important discrepancy is observed for exchange over a dry canopy during the day, when net emission is modelled in contrast with the observed average deposition velocity of 0.01 m s^{-1}. From this table it is again obvious that canopy wetness is a dominating factor in the dry deposition of NH_3.

7.3.5 THE AEROSOL PROJECT

Introduction

Particle, which are responsible for the atmospheric load to ecosystems consist of aerosols such as sulphate, nitrate, chloride, ammonium and base cations such as calcium, magnesium, sodium and potassium. Deposition of particles containing SO_4^{2-}, NO_3^-, Cl^- and NH_4^+ might contribute to the potential acidification and eutrophication (nitrogen components) of ecosystems. Compared to gaseous deposition of acidifying compounds, particle deposition velocities and fluxes are usually found to be small. However, accurate knowledge on particle deposition is necessary for areas far from sources since this might be the major atmospheric pathway of deposition, together with wet deposition, and because the long-range transport of particles is determined by a combination of their deposition velocity and scavenging ratio. Furthermore, it is believed that the dry deposition velocity of small particles and, with this, the fluxes, is currently underestimated for very rough surfaces like forests (Wiman *et al.,* 1990; Erisman, 1992; 1993a). Current knowledge is therefore insufficient to give an adequate assessment of the dry deposition of particulate sulphur and nitrogen over the Netherlands and Europe. There is a need for quantification to evaluate critical load exceedances and abatement strategies for atmospheric pollution.

Base cation deposition may be of importance for nutrient cycling in soils and ecosystems and may also neutralise acid input. Base cation input is therefore important in the determination of critical loads and/or critical load exceedances. Ecosystems receiving a high atmospheric input of base cations have higher critical loads than those receiving smaller inputs (Hettelingh *et al.*, 1991). Base cations are usually found in the coarse fraction of ambient aerosols. Their deposition velocities are therefore large.

In several studies where throughfall fluxes are compared with atmospheric deposition estimates, large differences have been found (e.g. Ivens, 1990, Draaijers and Erisman, 1993; Erisman, 1993b, see also Chapter 6). Establishing a link between the two is useful because it provides a way to estimate soil loads from atmospheric deposition estimates on the one hand, and allows the use of the relatively simple and cheap throughfall method to determine atmospheric deposition on the other. The link between atmospheric deposition and soil loads is important because critical loads refer to soil loads and because atmospheric deposition estimates provide a link with emissions. Thus, if critical load exceedances are used to estimate emission reductions, the relation between atmospheric deposition and soil load should be known. It was suggested by Draaijers and Erisman (1993) that besides canopy exchange processes, aerosol and/or fog and cloud-water deposition might be important processes contributing to the observed differences between atmospheric deposition estimates and throughfall measurements.

This section will summarise the results of the project. It is based on subprojects executed by the participating institutes (Ruigrok *et al.*, 1994; Römer and Te Winkel, 1994; Hofschreuder *et al.*, 1994; Duyzer *et al.*, 1994; Draaijers *et al.*, 1994; Vermeulen *et al.*, 1994; Van Leeuwen *et al.*, 1994; Arends *et al.*, 1994). First, the outline of the measurements and the model's development is explained. Results of the measurements and the model's application are then presented, followed by a discussion on canopy exchange, fog-water input, modelling and experimental validation and a generalisation of results. Detailed descriptions of subprojects are reported by the participating institutes. The main emphasis in this section is on the integration of subproject results.

Experimental set-up

The Speuld location was equipped with two towers and measuring facilities (see Figure 7.20). One tower was used for gas deposition measurements (Zwart *et al.*, 1994; Erisman *et al.*, 1993b,c; 1994e; see section 7.3.4). During the experiments, meteorological data and dry deposition data for SO_2, NH_3 and NO_2 obtained from this tower were available from the concurrent project on trace gas deposition (Erisman *et al.*, 1993b,c; 1994d; Wyers *et al.*, 1993, see section 7.3.4). The second tower was used for the aerosol project only. The tower is 34 m high and has a rectangular cross-section of 2.0 m x 1.5 m. An overview of the experimental set-up is shown in Figure 7.20. The measurements will only be described briefly in this section. A summary of the measurements is given in Table 7.21. For detailed descriptions of the equipment, sampling, analytical methods, selection of data, etc., please refer to the literature cited.

Micrometeorological experiments

Gradient measurements Gradients were determined by concentration measurements of $(NH_4)_2SO_4$ and NH_4HSO_4 at three heights (22, 26 and 34 m) by ECN using thermodenuders (Wyers *et al.*, 1994). Concentrations of total SO_4^{2-} and NO_3^- were measured at four heights (23, 26, 30 and 34 m) by TNO using filterpacks (Duyzer *et al.*, 1994). Concentrations of Cu^{2+}, Ti^{3+}, Ca^{2+}, Mg^{2+}, Na^+ and K^+ were measured at three heights (21, 29 and 35 m) using tunnel samplers (total particle mass), PM-10 devices (MD-50 of 10 μm) and cyclones (fine particles, MD-50 of 2.5 μm) by WAU (Hofschreuder *et al.*, 1994).

Eddy correlation measurements During a number of fog events, high frequency measurements (20.8 Hz) were made of turbulent characteristics and the liquid water content of fog (Vermeulen *et al.*, 1994; Wyers *et al.*, 1994). To characterise the droplet distribution function additional measurements were made with a Forward Scattering Spectrometer Probe (FSSP). Liquid water content fluxes were averaged over 10-min intervals. During the entire event, fog water was sampled actively with a string collector and analysed in the lab for its chemical composition (Römer and Te Winkel, 1994). More details can be found in Vermeulen *et al.* (1994).

Two Active Scattering Aerosol Spectrometer Probes (ASASP-X, Particle Measuring Systems, Boulder, Colorado, USA) were mounted in the tower at 18- and 25-m height by UMIST (UK). The instrument at 18 m was used in conjunction with a Gill Solent ultrasonic anemometer and at 25 m with a Kaijo-Denki ultrasonic anemometer (Beswick *et al.*, 1994). An ASASP-X measures particle sizes in the range of 0.1–3.0 μm diameter, this being split into 31 size bins plus an oversize channel. Both probes sampled through 4-m tubes. The equipment set-up allowed use of both the eddy correlation and the gradient technique for determining particle V_d. Data was logged at a frequency of 10 Hz (Beswick *et al.*, 1994). Measurements reported here were made between 29 June and 1 July.

Accumulation experiments

Accumulation of ^{214}Pb The ^{214}Pb activity accumulated on needles and the corresponding ^{214}Pb activity in air were measured simultaneously over a three-hour period for several days in 1993-1994. The dry deposition velocity for ^{214}Pb can be obtained from these measurements; which are expected to be representative for submicron particles since ^{214}Pb activity is attached to these particles. The experiments, based on the method previously used by Bondietti *et al.* (1984), are described in Wyers *et al.* (1994).

Deposition Filter Method (DFM) At three heights above the canopy (21, 29 and 35 m) deposition plates (DFM) were mounted to measure the accumulation of Cu^{2+}, Ti^{3+}, Ca^{2+}, Mg^{2+}, Na^+ and K^+ (Hofschreuder *et al.*, 1994). This method uses a horizontal paper filter as depositing surface. The filter was placed in a petri-dish with a 3-mm high edge which, in turn, was placed on a plate with a diameter of 30 cm. The filter was protected against precipitation by a roof mounted to the plate. Accumulation of particles was measured in the week before analysing. Between February and May 1993, about 14 weekly average samples were collected.

Washing Douglas fir and artificial branches Another series of accumulation experiments was carried out with needles from natural and artificial (polythene) branches (Römer and Te Winkel, 1994). Both Douglas fir and artificial branches were used in order to determine the influence of canopy exchange on the observed fluxes. Four Douglas fir branches were selected from near the mast at 11, 15, 17 and 19 m. Artificial branches were mounted on the tower at three heights (15, 17 and 19 m). The artificial branches were tested in the lab for chemical inertness. The needle surface was estimated beforehand using twig characteristics. Furthermore, the branches were taken into the lab and examined after the last experiment. The material accumulated during dry periods of several days was rinsed from the branches using a polypropylene bag filled with demineralised water.

Precipitation and throughfall measurements Rainfall and throughfall were collected in the Speulder forest for 10 months (Van Leeuwen *et al.*, 1994). Rainfall was measured with a wet-only device placed on top of the mast (35 m) and with four funnels in a clearing approximately 300 m from the mast. Two of the funnels were used for the mechanical sequential sampler. This sampler collects every 23 ml of volume rainwater (equivalent to 0.36 mm precipitation). Bulk throughfall was measured in 25 PVC gutters placed at an angle of approximately 24° from the horizontal. The gutters were placed in a regular pattern of two rows within a square of 250 m^2. For the sake of comparison, throughfall was also measured with 25 open samplers placed next to the gutters. A wet-only gutter, similar in size and form, and developed by the ECN, was placed near the gutters to determine wet-only throughfall. Throughfall was sampled sequentially during several events with two gutters connected to a shielded Campbell tipping bucket rain gauge. The minimum recording quantity was 0.04 mm and the maximum was set at 0.12 mm, after which sampling took place. Bulk rainfall and throughfall was collected in 30 periods. Samples were analysed within 24 hours of sampling for NH_4^+, PO_4^{3-}, Cl^-, HCO_3^-, NO_3^-, SO_4^{2-}, Ca^{2+}, Mg^{2+}, Na^+, K^+ and H^+.

Ambient concentration measurements
In addition to the measurements used for estimating deposition, concentrations of SO_4^{2-}, NO_3^-, NH_4^+, Cl^-, Na^+, K^+, Ca^{2+} and Mg^{2+} were measured in two size classes as 24-h averages (< 2.5 μm and > 2.5 μm) during a period of nine months (Römer and Te Winkel, 1994). Concentrations of HNO_2, HNO_3, HCl, SO_4^{2-}, NO_3^-, and NH_4^+ (< 2.5 μm) were determined for several days with annular denuders (Mennen (unpublished data); Arends *et al.*, 1994; Hofschreuder *et al.*, 1994) and once every week as hourly averages with wet rotating denuders (Wyers *et al.*, 1994).

TABLE 7.21 An overview of experiments performed at the Speulder forest site to quantify particle dry deposition

Technique	Species	Size range (μm)	Time resolution	Sampling heights (m)	Successful measurements	Random error flux (%)	Reference
Gradient measurements:							
Filterpacks	NO_3/SO_4	< ?[a]	2 h	24, 30, 35	39	75	Duyzer *et al.* (1994)
Thermodenuders	$(NH_4)_2SO_4$	< ?[a]	30 min	22, 26, 34	202	100	Wyers *et al.* (1994)
Cyclone, PM-10 tunnel sampler	Na, K, Ca, Mg	<2.5; <10; total	48 - 72 h	21, 29, 35	30	-	Hofschreuder *et al.* (1994)
Eddy correlation:							
Gerber	LWC	3 - 45	1 s	28	1637	20	Wyers *et al.* (1994)
FSSP	Fog droplets	1 - 95	15 min	28	1637	50	Wyers *et al.* (1994)
ASASP-x	Particles	0.1 - 3	0.1 s	18, 25	?[a]	50 - 150	Beswick *et al.* (1994)
Accumulation experiments:							
Accumulation on branches	^{214}Pb	< 1	3 h	18	26	60	Wyers *et al.* (1994)
Washing of branches	SO_4 ... Mg	0 - ∞	days	11, 15, 17, 19	13	60	Römer and Te Winkel (1994)
DFM	Na, K, Ca, Mg	0 - ∞	7 days	21, 29, 35	14	>25	Hofschreuder *et al.* (1994)
Throughfall	SO_4 ... Mg	0 - ∞	max. 1 week	1.5	30	30	Van Leeuwen *et al.* (1994)
Concentrations measurements:							
Wet denuder	HNO_3, HNO_2, HCl, SO_2	-	1 h	24	1000	15	Wyers *et al.* (1994)
ADS	SO_4 ... Mg	< 2.5	24 h	34	35	40	Hofschreuder *et al.* (1994); Mennen (unpublished data)
	HNO_3, HNO_2, HCl, SO_2		12 h	21, 34	16	30	
Aerosol sampler	SO_4 ... Mg	<2.5, 2.5-15	24 h	26	~280	5	Römer and Te Winkel (1994)

[a] unknown or no estimate available

Experimental determination of the acidifying aerosol and base cation input onto Speulder forest

In this section experimental deposition estimates of acidifying aerosol and base cations onto the Speulder forest will be summarised. The results are described extensively in Erisman *et al.* (1994d) and in a series of publications in *Atmospheric Environment* (Erisman *et al.*, 1995; Hofschreuder *et al.*, 1995; Ruigrok *et al.*, 1995; Wyers and Duyzer, 1995; Draaijers *et al.*, 1995; Gallagher *et al.*, 1995). The project aimed at validation of different processes in the particle deposition model. Results of this validation are described in section 7.3.5. In this section an outline of the assessment of aerosol deposition based purely on the measurements will be attempted.

As was previously known, it is obvious from the experimental results that determining particle deposition with measurements is very difficult. Even the most direct measurement technique, the eddy correlation measurements, sometimes shows controversial results with a large uncertainty. The most important factors leading to uncertainty are the measuring errors, the uncertainty in representativeness of measurement size distribution, the scaling factors for accumulation experiments, the influence of chemical conversion and the influence of humidity on particle growth. In general, the results show a distinct influence of the size distribution and u_* on the dry deposition velocity of particles. Figure 7.27 shows the relation between V_d and u_* for different experiments. The results are representative for different size distributions; see Table 7.22 (next section), showing V_d (^{214}Pb) < V_d (SO_4) < V_d (NO_3) < V_d (fog). V_d of fog is proportional to u_*^2, indicating that impaction is the most important process determining V_d. Sedimentation is also important. V_d values of other compounds are proportional to u_* or a weak function of u_*^x, with $0 < x < 1$, indicating no distinct dominating process but rather a mixture of processes occurring simultaneously (Van Aalst, 1986).

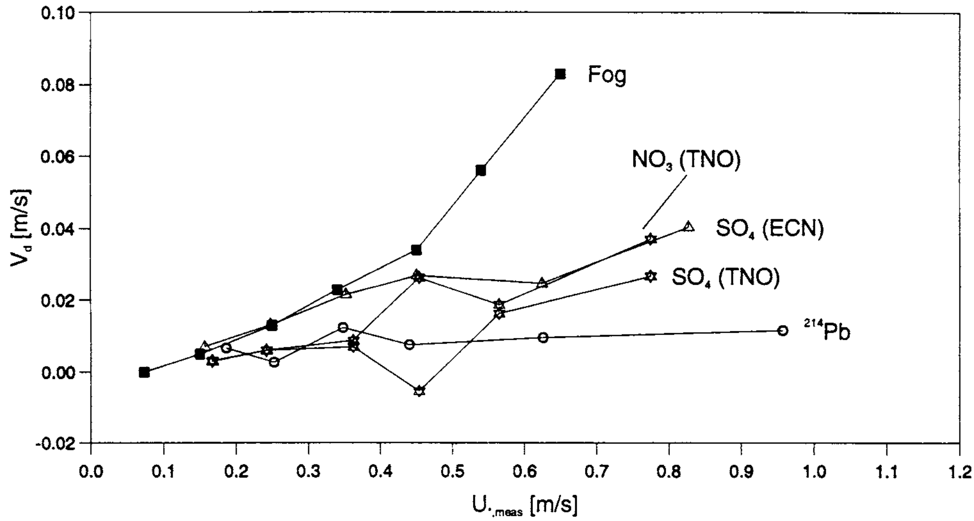


FIGURE 7.27 Experimentally determined V_d versus u_*. V_d values were estimated at the following heights: fog at 28 m; NO_3^- and SO_4^{2-} (TNO) at 35 m; SO_4^{2-} (ECN) at 22 m and ^{214}Pb at 19 m.

Results are in line with other investigations, showing that particle deposition to forests can be considerable, with dry deposition velocities of several cm s^{-1} (Hicks *et al.*, 1982; Waraghai and Gravenhorst, 1989; Sievering *et al.*, 1994). The relationships with u_* found for different particle size classes provide confidence in the experimental results. This can only be considered as an indication as errors in deposition estimates usually show a high correlation with u_*; this is because factors influencing the error in the deposition estimates are highly correlated to u_*.

Modelling particle deposition

A separate project dealing with modelling particle deposition was executed by KEMA (Ruijgrok *et al.*, 1994). First, the selection of the model and the sensitivity analysis for this project will be briefly described. Next, the results of the model evaluation with experimental results will be described and, finally, the suggested parametrisations will be discussed.

TABLE 7.22 Characteristic features of some models describing the dry deposition of particles to vegetation and water surfaces (Ruijgrok *et al.*, 1994)

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

A	Analytical model	+	Mechanism included
N	Numerical model	-	Mechanism not included
A/I	Analytical with iterative solving for some parts	and	Mechanism included by empirical data
		+/-	Mechanism considered, not included
		•	Neutral conditions only

Selection of models

A number of process-oriented particle deposition models were considered for an intercomparison study of models to measurement. The report by Ruijgrok *et al.* (1994) presents an overview of the models currently available for particle deposition. They consider two types of models, i.e. process-oriented and bulk-resistance models. The main characteristics of different process-oriented models are given in Table 7.22.

Bulk resistance models describe deposition in terms of resistances or deposition velocities applied over the entire size range of depositing particles. The EMEP model uses a uniform value for V_d of 0.1 cm s^{-1} (Iversen *et al.*, 1991). In the TREND model the resistance analogy is used with an adapted value for R_c. Voldner *et al.* (1986) used the same method; however, in their model R_b+R_c values were assumed to differ for receptor surfaces, seasons and day and night. Wesely *et al.* (1985) derived a parametrisation for V_d on u_*. This relation was modified by Erisman (1993a) and used for mapping particle deposition in the Netherlands. The results of these different methods display large differences in V_d (Ruijgrok *et al.*, 1994).

Current knowledge on the dry deposition process of particles is mainly qualitative. Only a small number of direct comparisons of particle deposition models with experimental data have been made. Results of most models, both process-oriented and bulk-resistance, indicate low values of V_d for small particles. This also applies to the dry deposition over very rough surfaces such as forests. Current process-oriented models for particle deposition require too many (unknown) parameters and are too uncertain for mapping dry deposition over large areas. On the other hand, no well-established and validated bulk resistance model exists for particle deposition.

From these models, the Slinn model (1982) was selected based on following criteria: it had to include the main processes with fundamental process descriptions and vegetation characteristics. Furthermore, it had to be easy to implement. In order to improve the model, four additions were made to the model, i.e.: 1) inclusion of growth in particle size at high relative humidity; 2) gravitational settling and impaction to include the Cunningham slip correction factor; 3) correction factors in the flux profile relations to account for deviations found from the original Dyer and Hicks (1970) relationships above the Speulder forest (Bosveld, 1991; Westrate and Duyzer, 1994); and 4) along with size-specific deposition velocities, an integration of V_d over the size distribution function, assuming values for *MMD* and the geometric standard deviation.

The uncertainty in model results was investigated using a Monte Carlo method (Janssen *et al.*, 1993). It was found that model results exhibit a large sensitivity to input conditions, especially for modelled V_d in the submicron size range. The upper limit of the central 95% of variation around the median V_d is several orders of magnitude larger than the lower limit. V_d for acidifying aerosols, integrated over the entire particle size distribution, was found to range from 1 mm s^{-1} to 5 cm s^{-1}. This range reflects natural variability (wind speed and roughness length) and uncertainty. For alkaline particles, in which most of the aerosol mass is concentrated between 1 and 20 μm, sensitivity to wind speed and roughness length is much smaller than that for acidifying aerosols. However, ranges in V_d are still substantial (Ruijgrok *et al.*, 1994).

Validation of the model using experiments

In order to simulate experimental results, the forest characteristics of the Speulder forest were estimated as accurately as possible. Ruijgrok *et al.* (1994) list the model input parameters used. Meteorological parameters such as wind speed, wind direction, Monin-Obukhov length, temperature and relative humidity were directly derived from measurements made in the Speulder forest. Model simulations were made for exactly the same times as for the micrometeorological experiments, and for the same measuring levels (V_d). For the results of accumulation experiments, averages over longer periods were calculated. To assess the model performance, the correspondence between modelled and measured values (V_d or F) is expressed in four measures: the correlation coefficient (R), the fractional bias (FBM) of mean observed and calculated values, (FBV) of the variances of observed and calculated values and the normalised mean square error ($NMSE$), which is a standardised measure of deviation between observed and calculated values (Ruigrok *et al.*, 1994). These measures are further explained in Ruijgrok *et al.* (1994). The values of these measures for the different comparisons are summarised in Table 7.24. In this table, averages, standard deviations and test results showing significant differences are also given.

Size distributions for particles were determined using values from the literature, measurements of the distribution of ^{214}Pb (Wyers *et al.*, 1994), measurements of alkaline particles in three classes (Hofschreuder *et al.*, 1995), measurements of 24-h average particle concentrations in two classes (Römer and Te winkel, 1994) and from a dataset of the size distribution of acidifying aerosols in 1983 and 1984 (Wyers *et al.*, 1994). The distributions thus obtained and used by Ruijgrok *et al.* (1994) are listed in Table 7.23.
For each experiment model calculations were made using the modified Slinn (1982) model with forests characteristics estimated for the Speulder forest, size distributions from Table 7.23, actual meteorology and concentrations measured above the forest, and the exact periods the experiments lasted. The individual model results are compared to measurements in (Ruijgrok *et al.*, 1994).

The experimental work was aimed at evaluating different aspects or process descriptions of the model. Most processes in particle deposition differ in their dependency on particle size. The best way to evaluate individual processes is to use measurements representative of different particle size classes. As discussed in Ruijgrok *et al.* (1994), the size distribution for which experiments are representative are uncertain. Furthermore, uncertainty in measurement results is usually so large that it is difficult to draw conclusions from a comparison. The experiments done here for evaluation of the modelled deposition mechanisms can be divided into four categories:

- ^{214}Pb measurements and eddy correlation measurements of particle deposition representative for small particles with diameters smaller than 1 μm
- NO_3^- and SO_4^{2-} measurements representative for particles with a bimodal distribution, with most of the total mass under 1 μm
- measurements of total base cation deposition representative for large particles
- fog deposition measurements representative for large particles.

Even though the *MMD* for which the four categories are representative differ considerably, none of these is representative for a single deposition mechanism. All four are determined mainly by impaction and interception. Sedimentation may play an important role in fog deposition, depending on the size of droplets.

TABLE 7.23 Component specific-size distributions (mass median diameter, *MMD*, and geometrical standard deviation, σ_g) derived from measurements in the Netherlands

Component	*MMD* (μm)	σ_g
^{214}Pb	0.35	2.0
Ca^{2+}	7.73	3.47
Mg^{2+}	5.92	2.73
K^+	2.64	1.84
Na^+	5.12	2.64
SO_4^{2-} < 2.5 μm	0.6	2.2
SO_4^{2-} > 2.5 μm	4.5	1.6
NH_4^+ < 2.5 μm	0.6	2.2
NH_4^+ > 2.5 μm	4.0	1.6
NO_3^- < 2.5 μm	0.6	2.3
NO_3^- > 2.5 μm	4.5	1.6
Fog droplets (December)	19.4	
Fog droplets (February)	7.4	

The next factor which is important for particle deposition is the friction velocity. Similar size dependencies and u_* dependence of the dry-particle deposition velocity for the measurements and the model results, might serve as some sort of validation of the most determining processes. However, one has to be careful in drawing firm conclusions from such a comparison. Errors in deposition estimates usually show a high correlation with u_*, because factors influencing the error in the deposition estimates are highly correlated to u_*, e.g. the stability corrected gradient. For the u_* dependence of V_d, Figures 7.27 and 7.28 need to be compared. In these figures, the measured or modelled V_d is averaged for each class of u_*. For base cations, the u_* dependence cannot be compared because only total fluxes have been compared. The figures show that the three other categories distinguished above show different dependencies on u_*, as might be expected. Furthermore, the figures show similar relations between V_d and u_* for the three categories, although the variation in measured V_d per u_* class can be very high.

Evaluation of the integral model results can only be done with statistical parameters, as displayed in Table 7.24, when the uncertainty in modelled and measured values is taken into account. The uncertainty in model results and the sensitivity of model results on input and model parameters has been extensively described in Ruijgrok *et al.* (1994). The overall random error in modelled V_d integrated over the size distribution representative for acidifying aerosols (Table 7.23) is estimated at about 65% (Ruijgrok *et al.,* 1994). For base cations this error is somewhat smaller (60%) because of the contribution of the relatively well-parameterised description of sedimentation. The uncertainty in model estimates is lower than or about equal to the uncertainty in measurement results, with the exception of the fog deposition measurements, which are estimated to have smaller errors (20%). The fractional bias of the means (the relative difference between the mean calculated and observed values) falls within these limits (Table 7.24). The relatively large sensitivity of the model and an inaccuracy of the same order of magnitude in measuring results mean that a perfect 1:1 correspondence cannot be expected. Statistical testing of the difference between modelled and measured values is done using the non-paramatric sign and Wilcoxon tests for paired samples (Table 7.24). Both tests revealed no significant differences between the mean values of modelled and measured fluxes or V_d*'s* (Ruijgrok *et al.*, 1994). This is of course mainly the result of the large standard deviations in measuring and modelling results.

TABLE 7.24 Model performance indicators[a] and averages for the different comparison studies

Intercomparison	*R*	*FBM*	*FBV*	*NMSE*	V_d model	σ model	V_d meas.	σ meas.	*n*	Remarks[b]
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^- (filterpacks)	0.55	0.02	-1.06	1.75	1.2	1.1	1.2	1.9	23	
SO_4^{2-} (filterpacks)	0.42	0.55	-0.60	2.66	1.1	1.0	0.7	1.4	23	
SO_4^{2-} (thermodenuders)	0.33	-0.08	-1.54	1.94	2.1	1.2	2.3	3.4	169	
Ca^{2+} (DFM, scaled)	0.78	0.62	0.93	0.59	4.1	2.5	2.4	0.9	14	

[a] Perfect agreement if R=1, FBM=0, FBV=0, $NMSE$=0; no agreement if R=0, $|FBM|$=2, $|FBV|$=2, $|NMSE|=\infty$

[b] Non-significant differences between mean calculated and measured values of V_d (cm s^{-1}) are denoted with ** (p<0.05 in a Wilcoxon paired sample test); Non-significant (<95%) correlation coefficients (R) are denoted with and.

In conclusion, there are no strong indications for a significant underestimation or overestimation of the modelled V_d compared to measured values. The fact that the model is reasonably capable of describing a response of V_d to u_* similar to the measurements for different particle diameters provides confidence in the process descriptions.

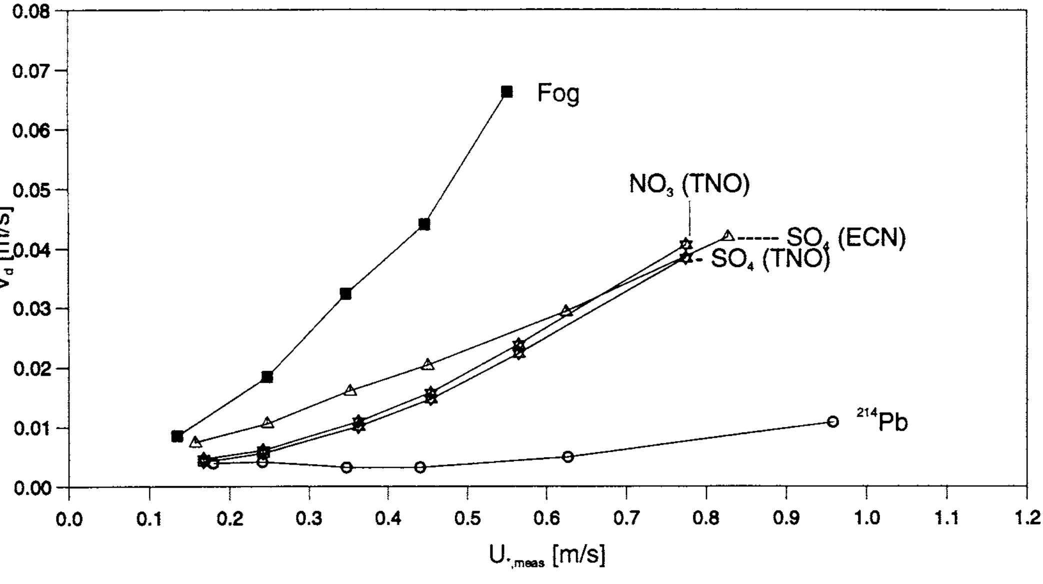


FIGURE 7.28 Summary of modelled deposition velocities versus u_*. V_d values were estimated at the following heights: fog at 28 m; NO_3^- and SO_4^{2-} (TNO) at 35 m; SO_4^{2-} (ECN) at 22 m and ^{214}Pb at 19 m.

Validation of the model using accumulation experiments

Long-term experiments were executed to evaluate the balance of measured and modelled fluxes. It has been shown that most of the experiments at Speulder forest point in a similar direction regarding canopy exchange. Although the estimates of the absolute amounts of components retained or leached in the canopy may differ depending on experiment, the average values (Table 7.25) give a good picture of the situation at Speulder forest. The table shows that H^+ is taken up by the canopy, which is accompanied by leaching of Mg^{2+}, Ca^{2+} and most of all K^+. SO_2 taken up by stomata is eventually leached again, whereas NH_3 taken up via stomata is almost completely retained by the canopy. Oxidised nitrogen components are taken up by the canopy, especially NO_2. Whether NO_3^- and NH_4^+ aerosols are taken up directly or via solution is uncertain. We assume from the results of the experiments that NO_3^- uptake is negligibly small, whereas NH_4^+ is taken up in exchange for K^+ or Ca^{2+}. Na^+ and Cl^- are considered inert. The highest uncertainty in canopy exchange estimates relates to estimates of the nitrogen components and Ca^{2+} and Mg^{2+}.

Observed differences between dry and fog deposition estimates from micrometeorological measurements and inferential modelling on the one hand and net throughfall fluxes on the other cannot be regarded exclusively due to canopy exchange. Dry deposition estimates from micrometeorological measurements and inferential modelling are uncertain through errors in the air concentration measurements (Arends *et al.*, (1994), their sometimes low time coverage and the uncertainties associated with the parametrisation of the dry deposition velocities (Erisman *et al.*, 1994; Ruijgrok *et al*, 1994). Fog deposition estimates are uncertain due to uncertainties associated with the estimation of water fluxes and the measurement of the average chemical composition of the fog droplets (Vermeulen *et al.*, 1994). Uncertainties associated with the throughfall method when used for estimating dry and fog deposition include the dry deposition to the forest floor and understorey vegetation, dry deposition directly onto the throughfall gutters, the representativeness of the throughfall sampling, the wet deposition estimate, the stemflow contribution and canopy exchange processes (Draaijers and Erisman, 1993). With canopy exchange processes being the only exception, aforementioned factors probably contributed only to a very small extent to the uncertainty in the throughfall dry and fog deposition estimates in this study.

TABLE 7.25 Average uptake and leaching amounts at the Speulder forest derived from field experiments and modelling (mol ha^{-1} a^{-1}) (Draaijers *et al.*, 1994)

Component	Uptake	Leaching
SO_2	35	0
SO_4^{2-}	0	80
NO_2	115	0
HNO_2	15	0
HNO_3	0	0
NO_3^-	0	0
NH_3	140	0
NH_4^+	115	0
Na^+	0	0
Cl^-	0	0
Mg^{2+}	0	40
Ca^{2+}	0	50-75
K^+	0	250
H^+	190	0

7.3.6 ANNUAL AVERAGE GAS AND PARTICLE DEPOSITION AND CHANGES WITH TIME

Meteorological data and dry deposition data for SO_2, NH_3 and NO_2 were available from the continuous gradient and eddy correlation measurements for quantifying the input of acidifying gases and aerosols, and base cations, onto the Speulder forest (section 7.3.4, Erisman *et al.*, 1993a,b,d; Wyers *et al.*, 1993). Annual averages were determined by averaging the dry deposition parameters for the selected periods and those derived by the surface resistance parametrisation, concentration and meteorological data for the rejected periods. Particle input of acidifying aerosols and base cations, and fog-water deposition were estimated using measurements of concentrations, meteorological parameters and model results (section 7.3.5, Erisman *et al.*, 1994). Wet deposition is estimated using interpolated values from the National Air Quality Monitoring Network (e.g. RIVM, 1993). Table 7.26 summarises the results of the experiments and model calculations regarding fluxes of different compounds. In this table measurements and flux estimates made at the Speulder forest site during the first and second phase of the Dutch Programme on Acidification are also listed. These data have been taken from Van Aalst and Erisman (1991) and Duyzer *et al.* (1994). The estimates of the canopy uptake reflect the needle uptake of gases (SO_2, NH_3 and NO_2) through stomata and the uptake through cuticles from the water layer covering the needle surface. This is not the net uptake, because part of this flux might be leached or evaporated again. Above-ground uptake and leaching by the canopy (Table 7.25) was estimated by Draaijers *et al.* (1994) by *i)* comparing atmospheric deposition estimates with throughfall fluxes, *ii)* applying several canopy budget models; *iii)* comparing deposition estimates from surface wash experiments using real and artificial Douglas fir twigs and *iv)* using results from a S^{35} nutrition experiment. Stomatal uptake of gases was estimated from concentration measurements and the stomatal conductance model of Bosveld and Bouten (1992).

TABLE 7.26 Average fluxes at Speulder forest in 1987-1993 (mol ha^{-1} a^{-1})

Compound	Year	Measurements and inferential modelling	Throughfall measurements[b]	Canopy uptake	Canopy leaching
SOx:	1987		1180		
(SO_2, SO_4^{2-}, wet SO_4^{2-})	1988	1220[c]	1120		
	1989	980[c]	1020		
	1992/1993	1180	1090	35	80
NHx:	1987		2870		
(NH_3, NH_4^+, wet NH_4^+)	1988	2975[c]	2740		
	1989	2910[c]	2630		
	1992/1993	2865[a]	2560	255	0
NOy:	1987		820		
(NO_2, HNO_2 , HNO_3,	1988	1105[c]	720		
NO_3^-, wet NO_3^-)	1989	1235[c]	820		
	1992/1993	1080	770	130	0
Cl^-	1987		1060		
	1988		1640		
	1989		1090		
	1992/1993	1425	1290	0	0
Na^+	1987		850		
	1988		1160		
	1989		770		
	1992/1993	1065	1040	0	0
Ca^{2+}	1987		380		
	1988		280		
	1989		250		
	1992/1993	145	200	0	50-75
Mg^{2+}	1987		240		
	1988		310		
	1989		220		
	1992/1993	190	270	0	0-40
K^+	1987		70		
	1988		40		
	1989		40		
	1992/1993	75	80	0	270

[a] Net-flux, emissions accounted for (see text)
[b] Data are in very good agreement with KUN measurements (Boxman, 1994, personal communication). Throughfall is corrected for canopy exchange with a model as described by Draaijers *et al.* (1994).
[c] Data derived from Van Aalst and Erisman (1991); particle fluxes corrected for the underestimated V_d of particles (see section 7.3.5).

Measurements of atmospheric loads and throughfall for the years before 1987 are not available. Deposition estimates for the acidifying components for the years between 1980 and 1987 are available from the DEADM model (Chapter 5, Erisman, 1993a) and presented in Table 7.27, along with estimates for the deposition before 1980. These are based on the data for 1980, scaled by the ratio of the deposition in the Netherlands between 1950 and 1980 estimated by the DAS model on the basis of emissions in these years (De Boer and Thomas, 1991). Deposition estimates for base cations are not available for the years before 1993.

TABLE 7.27 Estimates of the deposition of acidifying components between 1950 and 1994 for the Speulder forest site (mol ha^{-1} a^{-1})

Year	SOx	NHx	NOy	Total nitrogen	Potential acid
1950	1700	1350	740	2090	5490
1955	2230	1410	770	2180	6640
1960	2320	1600	870	2470	7110
1965	3100	1790	980	2770	8970
1970	2710	2180	1190	3370	8790
1975	2030	2630	1440	4070	8130
1980	1940	2690	1470	4160	8040
1981	1940	2560	1600	4160	8040
1982	1700	2580	1540	4120	7520
1983	1510	2780	1560	4340	7360
1984	1590	2690	1580	4270	7450
1985	1530	2740	1560	4300	7360
1986	1570	2600	1470	4070	7210
1987	1190	2970	1490	4460	6840
1988	1220	2980	1110	4080	6520
1989	980	2910	1240	4150	6110
1990	1170	2830	1220	4050	6390
1991	1080	2820	1190	4010	6170
1992	1080	2890	1240	4130	6290
1993	1040	2740	1200	3940	6020

From Table 7.27 it is obvious that the highest input of potential acid was in the seventies. After 1965 the acid input gradually decreased from 9000 to 6000 mol ha^{-1} a^{-1}. The nitrogen input increased from 1950 to 1987. In 1987 a maximum nitrogen input of 4500 mol ha^{-1} a^{-1} (63 kg ha^{-1}) was reached. After 1987 the nitrogen input decreased to 3900 mol ha^{-1} a^{-1} (~55 kg ha^{-1}). Throughfall measurements of NH_4^+ and NO_3^- made between 1988 and 1993 show a slight decrease in soil N load (Table 7.26). The main contribution to the nitrogen input is formed by ammonia and ammonium (70%). Throughfall fluxes of base cations show strong year-to-year variation. Base cation input is mainly the result of dust emissions from nearby roads and emissions due to industrial and agricultural activities. The input of sodium is determined by sea salt deposition. High input occurs during periods with high wind speeds from a westerly or south-westerly direction.

7.3.7 ASSESSMENT OF THE EFFECTS OF ACIDIFICATION, EUTROPHICATION AND OZONE

Critical levels and loads at the Speulder forest site

Generally, critical levels are expressed in terms of exposure ($\mu g\ m^{-3}$ and exposure duration) while critical loads are expressed in terms of deposition (mol $ha^{-1}\ a^{-1}$ or kg $ha^{-1}\ a^{-1}$). Critical levels focus on no-effect thresholds for short-term exposures (one year or less) and critical loads focus on safe deposition quantities for the long term (10-100 years). Critical levels and loads are not aimed to completely protect vegetation against adverse effects. No-effect levels and loads are usually higher and lower than the critical level or loads, respectively (Steingröver *et al.*, 1995). Exceedances are determined by estimating the difference between actual levels and loads, and critical levels and loads, for the time and scales that the critical values were estimated.

Critical levels

In order to assess critical levels, information is needed on quantitative relationships between exposure and effect. In many cases this information is not available or based on only few experiments and therefore rather speculative. Specific critical levels for the Speulder forest can not be assessed with current knowledge.

SO_2 In order to prevent visual damage (leaf discoloration and/or loss) to coniferous and deciduous forests on poor sandy soils the annual, daily and hourly average SO_2 concentration should not be higher than 25 $\mu g\ m^{-3}$, 70 $\mu g\ m^{-3}$ and 150 $\mu g\ m^{-3}$, respectively (Heij and Schneider, 1991; Table 7.28). Van der Eerden (1995) mentions a critical level of 20 $\mu g\ m^{-3}$ (winter and annual mean).

NOx To prevent effects on photosynthesis and growth, for example, the critical level for NO_x (NO and NO_2, added in ppb and expressed as NO_2 in $\mu g\ m^{-3}$) is set to 30 $\mu g\ m^{-3}$ as an annual mean, and 75 $\mu g\ m^{-3}$ as a 24-h mean (Van der Eerden *et al.*, 1995). Interactive effects between NO_2 and O_3 and/or SO_2 have been reported frequently, but the lowest effective levels for NO_2 are about equal to those for combination effects. Generally, at concentrations near to its effect threshold, NO_2 causes growth stimulation if NO_2 is the only pollutant, while in combination with SO_2 and/or O_3 it results in growth inhibition (Van der Eerden, 1995).

NH_3 Exposure to high NH_3 concentrations (>180 $\mu g\ m^{-3}$) or high NH_4^+ concentrations in canopy surface water (>5 mmol l^{-1}) is found to damage the crystalline structure of the epicuticular wax layer of the needles of Douglas fir (Van der Eerden *et al.*, 1992). Critical levels for adverse effects of NH_3 on plants were estimated by Van der Eerden *et al.* (1991). To protect 95% of the species at $p<0.05$ a 24-h and annual mean of 270 and 8 $\mu g\ m^{-3}$, respectively, was estimated.

O_3 Recently, a threshold value for O_3 for forest ecosystems was set at 40 ppb. As a provisional critical level, an *AOT* (Accumulative exposure Over a Threshold concentration) has been defined. The *AOT40* represents the summed hourly concentrations in the growing season (April-September) with concentrations above 40 ppb (Fuhrer and Achermann, 1994). To protect 95% of the tree species against a growth reduction of more than 10%, the AOT40 should be lower than 10 ppm h (Van der Eerden, 1995). This *AOT40* value was derived from O_3 exposure experiments using juvenile trees in open-top chambers.

TABLE 7.28 Critical levels for forests in Europe

Gas	Effects	Critical level	Reference
SO_2	Visual damage	yearly average: 20-25 μg m^{-3} daily average: 70 μg m^{-3} hourly average : 150 μg m^{-3}	Schneider and Bresser (1988); Van der Eerden (1995)
NO_x	Photosynthesis and growth reduction	annual average: 30 μg m^{-3} daily average: 75 μg m^{-3}	Van der Eerden *et al.* (1994)
NH_3	95% of the species protected	hourly average: 3300 μg m^{-3} daily average: 270 μg m^{-3} monthly average: 23 μg m^{-3} yearly average: 8 μg m^{-3}	Van der Eerden *et al.* (1991)
	Damage to epicuticilar wax layer	NH_3 in air > 180 μg m^{-3} NH_4^+ in water layer > 5 mmol l^{-1}	Van der Eerden *et al.* (1992)
O_3	Biomass reduction > 10%	AOT40 (growing season) = 10 ppm h	Fuhrer and Achermann (1994)

Critical loads

Critical loads for acidity for the Speulder forest were derived using the steady-state mass balance model of De Vries (1993). The equations, basic assumptions and values used to estimate the critical loads are given in Erisman *et al.* (1995). It is of importance to notice that the newest insights are used to estimate the critical loads of acidity, i.e. a critical $Al^{3+}/(Ca^{2+} + Mg^{2+} + K^+)$ ratio of 1.0 mol mol^{-1} for coniferous forest and 3.3 mol mol^{-1} for Douglas fir is used instead of the usually used Al^{3+}/Ca^{2+} ratio of 1.5 mol mol^{-1}. These ratios are also used for the root damage criteria instead of a critical Al^{3+} concentration. The critical load values of acidity for the Speulder forest are listed in Table 7.29.

Critical acid loads for the Speulder forest are about 100% larger than average critical acid loads derived for coniferous forests on sandy soils in the Netherlands found by de Vries and Kros (1991) because of *i)* the different criteria used: $Al^{3+}/(Ca^{2+}+Mg^{2+}+K^+)$ ratio instead of Al^{3+}/Ca^{2+} ratio, and Al^{3+} concentration, *ii)* the high weathering rate at Speulder forest and *iii)* the high values for N uptake (actually, one should use values at critical N loads and not the current value).

TABLE 7.29 Critical acid loads for Speulder forest based on different effects (mol ha^{-1} a^{-1})

Effects	Criteria	Critical load
Inhibition of uptake/root damage	$Al^{3+}/(Ca^{2+} + Mg^{2+} + K^{+}) = 1.0$ mol mol^{-1}	2760[a]
	$Al^{3+}/(Ca^{2+} + Mg^{2+} + K^{+}) = 3.3$ mol mol^{-1}	5990
Al^{3+} depletion	$\delta\ Al(OH)_3 = 0$ mol kg^{-1}	2725
Al^{3+} pollution	$Al^{3+} = 0.02$ mol m^{-3}	915

[a] Using a critical molar Al^{3+}/Ca^{2+} ratio of 1.0 yields a value of 1860 mol ha^{-1} a^{-1}. A critical Al^{3+} concentration, often used in critical load calculations to date, leads to a critical acid load of 1970 mol ha^{-1} a^{-1}.

The critical N loads for the Speulder forest related to vegetation changes, increased sensitivity and NO_3 pollution of the groundwater were also derived with the steady-state mass balance model according to De Vries (1993). The derivation of the critical N loads is also given in De Vries (1993) and for Speulder forest in Erisman *et al.* (1995). Critical loads for N are listed in Table 7.30.

TABLE 7.30 Critical N loads for Speulder forest based on different effects (mol ha^{-1} a^{-1})

Effects	Criteria	Critical load
Vegetation changes	N = 0.1 mol m^{-3}	810
Increased sensitivity	N = 1.8%	1500
NO_3^- pollution	$NO_3^- = 0.4\text{-}0.8$ mol m^{-3}	1950-3190
Inhibition of uptake[a]	$NH_4^+/K^+ = 5$ mol mol^{-1}	2460

[a] Refers to NH_3 only

The critical load for ammonia, related to the occurrence of nutrient imbalances in the soil solution, was also calculated with a steady-state model according to De Vries (1993). This critical load is also listed in Table 7.30. The critical NH_3 load derived for the Speulder forest is approximately three times as large as the lower value given by Bobbink *et al.* (1992) for coniferous forest growing on non-calcareous sandy soils in the Netherlands (800 mol ha^{-1} a^{-1}). The latter value was derived in an experiment with trees with elevated N/K ratios in needles, as compared to those at critical loads, and a soil with very low (negligible) nitrification rate. For soils with increased cation rates, Bobbink *et al.* (1994) reports values up to 4000 mol ha^{-1} a^{-1}.

Uncertainties in critical loads are due to uncertainties in the calculation method, the critical chemical values and criteria used, and the input data. The largest uncertainties in critical (acid) loads are likely to be due to the uncertainties in critical chemical values. Whether one uses a critical $Al^{3+}/(Ca^{2+}+Mg^{2+}+K^{+})$ ratio of 1.0 (for coniferous forest) or 3.3 mol mol^{-1} (for Douglas fir) will change the critical acid load from less than 3000 to about 6000 mol ha^{-1} a^{-1}. Furthermore, the uncertainties in input data may also lead to large uncertainties in the critical loads. The estimated weathering rate at the Speulder forest, for example, is based on an input - output budget and includes cation exchange as well. Generally, average weathering rates for acid sandy soils are estimated at 200 mol ha^{-1} a^{-1}, which is much lower than the value of 550 mol ha^{-1} a^{-1} used for Speulder forest. Furthermore, one has to be aware that the critical loads previously derived are all based on present data with respect to dry deposition, weathering, uptake, litterfall and throughfall. Actually one should use the values at critical loads. Elevated present N uptake rates cause an increase in critical loads for N and acidity, whereas elevated N/K ratios in needles decrease the critical load for NH_3 related to inhibition of uptake. In general, the uncertainty in criteria and input data may cause an uncertainty of more than 50% in most of the critical loads derived.

Effect parameters and observed effects

What are the effect parameters?
Before answering the question on what the observed effects are, we need to define what we mean by effects and what kind of effect parameters are taken into account. Effects are defined as ecosystem changes as a result of environmental impacts/stress. In general, direct and indirect effects are distinguished. Direct effects are the result of exposure to air pollutants, whereas indirect effects are the result of soil loads and changes in soil solution. On the other hand, direct and indirect effects are sometimes distinguished according to the time between cause and observed effect (i.e. effects on photosynthesis versus effects on growth).

In practice, the health (vitality) of trees and of entire forest ecosystems is their capacity to cope with stress: not only the result of the occurrence of air pollution but also natural stresses caused by changes in environmental conditions (drought, frost, windthrow, pests, diseases, etc.). Usually, it is difficult to derive mono-causal relations from the multiple stress to which forests are exposed.

An overview of possible effects on forests as a result of increased atmospheric acid and nitrogen deposition and/or exposure to air pollutants is presented in Table 7.31. Based on the processes and effects mentioned in this table, three ecosystem compartments can be distinguished: the trees, the understorey and the soil and litter layer. In these compartments we can distinguish between several effect indicators or parameters, which will be used to relate changes to environmental stresses. For the trees we distinguish growth, photosynthesis, transpiration and water stress, the nutrient composition of needles and vitality; for the understorey changes in composition of flora and fauna, and for the soil and humus layer we distinguish parameters such as water and nutrient fluxes, root and mycorrhiza parameters. The increased atmospheric acid and nitrogen load and air pollutant concentrations lead to changes in the aforementioned effect parameters and/or changes in sensitivity and thus increase the risk of damage due, for example, to plagues, diseases, storms, drought and frost sensitivity.

The changes in the effect parameters (underlined in Table 7.31) in the Speulder forest during the research period are discussed and evaluated in Erisman *et al.* (1995). Here the outcome will only be used in a synthesis.

TABLE 7.31 Possible effects on forest ecosystems of increased atmospheric N+S loading and exposure to SO_2, NOx, NH_3 and/or O_3. Effect parameters are underlined.

Ecosystem compartment	Effects chemistry	Ecosystem
Trees (including foliage and roots)	proton accumulation in foliage	- damage of epicuticular wax layer of foliage
	O_3	- decrease of <u>photosynthesis</u> and respiration
	SO_2, O_3 , NH_3	- enhanced <u>transpiration</u> and drought sensitivity by elevated stomatal control
	elevated <u>N contents in foliage</u>	- increased frost sensitivity
		- increased parasite injury (insects, fungi, virus)
		- increased <u>ratio of foliage to roots</u> (risk of drought and nutrient deficiency)
		- increased <u>biomass production</u>
		- increased <u>water demand</u>
		- increased cell size in stems
	elevated <u>arginine concentration</u>	- <u>growth reduction</u>
	<u>nutrient deficiency</u> absolute or relative (to N)	- discoloration (<u>defoliation</u>)
Soil (solution)	elevated N contents in soil	- increase in nitrophilus species
		- decrease in biodiversity
	depletion of secondary Al compounds	- possible <u>root damage</u> by pH decrease and nutrient deficiencies
	<u>elevated concentrations</u> (leaching) of H + Al	- <u>root damage</u>
		- <u>mycorrhiza decline</u>
	<u>elevated ratios</u> of NH_4 and Al^{3+} to base cations	- inhibition of uptake (nutrient deficiency)

Synthesis

Exceedance of critical levels

At the Speulder forest critical levels for SO_2 were exceeded from 1955 up to 1975. In this period, annual average SO_2 concentrations were above 25 $\mu g\ m^{-3}$. From 1975 onwards, annual average SO_2 concentrations at the Speulder forest were lower than the critical level, and decreasing steadily. However, daily and hourly average critical levels of SO_2 may still have been exceeded during periods of winter smog. Annual average NOx concentrations at the Speulder forest have, since 1950, always been lower than the critical levels. As for SO_2, daily average critical levels of NOx may have been exceeded during smog periods. The critical levels for NH_3 probably have not been exceeded at the Speulder forest. The highest annual mean concentration at the forest was estimated to be 4 μg m^{-3} (Erisman *et al.*, 1995) whereas 24-h means never exceeded 10 μg m^{-3} (Wyers *et al.*, 1993; Vermetten *et al.*, 1991). Concentrations of NH_4^+ observed in sequentially sampled throughfall water at the Speulder forest were as high as 4 mmol l^{-1} (Van Leeuwen and Bleuten, 1994). As dilution of the water films covering the needle surface occurs during the wash-off process, it is likely that the foliage is exposed to larger concentrations than those measured in throughfall, thereby probably close to or even exceeding critical levels. The *AOT40* for O_3 has been exceeded since measurements started in 1979 and have probably been exceeded since planting the forest 33 years ago.

There is no hard evidence that the direct effects found at Speuld related to air pollutant exposure result in a significant decrease in vitality and or sustainability of the trees. Critical levels of O_3 have been exceeded continuously since O_3 measurements started in 1979. In 1992, total assimilation was reduced by 2.5%-7% at an *AOT40* of 39.1 ppm h. In 1993 the reduction found was approximately twice as large at an *AOT40* of 23.8 ppm h (Steingröver and Jans, 1995). Van der Eerden (1995) suggests that the *AOT40* of 10 ppm h set to protect tree species against a growth reduction of more than 10% may be too small. Mature trees under field conditions may have some kind of feedback mechanism through which they can cope with larger O_3 concentrations compared to juvenile trees in open-top chambers. Comparison of 3-, 17- and 33-year old trees at the Speulder forest revealed that the youngest trees had relatively more reactive stomata and thus were more sensitive to ozone and vapour pressure deficits (Steingröver and Jans, 1995). This is in agreement with findings of Colls *et al.* (1993) and Pleijel *et al.* (1993). No direct effects of SO_2 and NO_2 were observed. This agrees with the observation that critical levels of these components have not been exceeded. Critical levels for NH_3 have not been exceeded either. Occasionally, NH_3 was found to stimulate the net assimilation rate. Exposure to high O_3 and/or high NH_3 concentrations was suggested as the cause of stomata remaining open even at low shoot-water potentials and/or high vapour pressure deficits, through which trees become more vulnerable to drought. NH_4^+ concentrations in water layers covering the needle surface were sometimes (especially after long dry periods and/or during fog episodes) such that damage of the epicuticular wax layer was likely to occur. Such effects have, however, not been investigated at Speulder forest.

Exceedance of critical loads

The critical loads related to most effects are exceeded since planting the trees some 33 years ago. Table 7.32 gives information on the exceedances of the critical loads and whether the effects related to these exceedances have been observed at Speulder forest. Comparison of current acid loads (ca. 6000 mol ha^{-1} a^{-1}; Table 7.27) with critical acid loads (Table 7.29) shows that the critical loads are exceeded since the trees were planted, especially those related to the pollution of phreatic groundwater (see Table 7.32). The critical load for inhibition of uptake and root damage are exceeded since planting whenever both a critical $Al^{3+}/(Ca^{2+}+Mg^{2+}+K^{+})$ ration of 1.0 mol mol^{-1} is used and when the value for Douglas fir is used (3.3 mol mol^{-1}).

Comparison of current N loads (ca 3900 mol ha^{-1} a^{-1}) with critical N loads (Table 7.26) shows that NO_3^- pollution above the EC drinking-water standard may occur in the long term when N immobilisation becomes negligible. The exceedance of N loads related to vegetation changes is irrelevant for this production forest because it is a monoculture with no understorey vegetation. The critical load for increased sensitivity is exceeded since the trees were planted. The critical load for inhibition of uptake (NH_3) is also exceeded since the trees were palnted, but only by a few hundreds of moles. This is due to the relatively high nitrification rate at the Speulder forest. The present N input does, however, not preclude nutrient imbalances in the foliage. It may cause too high N contents related to Ca^{2+}, Mg^{2+} and/or K^+.

TABLE 7.32 Critical loads, exceedances and effects observed at Speulder forest

Effects	Criteria	Exceedance[b]	Observed effects at Speulder forest
Inhibition of uptake/root damage	$Al^{3+}/(Ca^{2+}+Mg^{2+}+K^+) = 1.0$ mol mol^{-1}	always	inhibition of base cation and phosphorus uptake; decrease in mycorrhiza
	$Al^{3+}/(Ca^{2+}+Mg^{2+}+K^+) = 3.3$ mol mol^{-1}	always	
Al^{3+} depletion	δ $Al(OH)_3 = 0$ mol kg^{-1}	always	aluminium depletion
Al^{3+} pollution	$Al^{3+} = 0.02$ mol m^{-3}	always	aluminium leaching
Vegetation changes	$N = 0.1$ mol m^{-3}	always	non, no understorey
Increased sensitivity to drought, storms, frost and fungal diseases	N = 1.8%	always	large growth stimulation; high foliage N concentration and N/K ratio (frost, fungal diseases); high foliage/fine root ratio high water demand; high foliage/coarse root ratio (-> storms)
NO_3^- pollution of groundwater	$NO_3^- = 0.4$-0.8 mol m^{-3}	always - 1970	decrease in mycorrhiza; nitrogen saturation; nitrate leaching
Inhibition of uptake[a]	$NH_4^+/K^+ = 5$ mol mol^{-1}	always	P, K , N/P and N/Mg deficient, increase in arginine

[a] Refers to NH_3 only
[b] Always refers to 'since the plantation of the forest in 1961'

There are several uncertainties associated with the estimates of exposure and loads, but also with the estimates of the critical levels and loads (De Vries, 1995; Erisman *et al.*, 1995). Furthermore, critical loads should evolve during time, which is not taken into account. This might all have an effect on the exceedance and thus the determination whether the forest is at risk or which effects are expected. The critical loads are defined in such a way that there is an increased risk when both the exceedance or the time of exceedance increases. The expected effects related to exceedance and exceedance time are thus related to Al^{3+} depletion and leaching, deviation from optimal growth, increased sensitivity to drought, storms, frost and fungal diseases, and to some extent to inhibition of uptake and root damage. Considering the changes in effect parameters, it can be concluded that the evolution of loads at the Speulder forest has already lead to *i)* a decrease in mycorrhiza through which base cation and phosphorus uptake is inhibited, leading to a relative base cation and phosphorus deficiency in the foliage, *ii)* a high foliage/fine root and foliage/coarse root ratio through which the forest becomes increasingly sensitive to drought and windthrow; *iii)* depletion of readily available Al^{3+} compounds, causing a further pH decline in the future (De Vries, 1993), and *iv)* leaching of Al^{3+} and NO_3^- to the groundwater. It can thus be concluded that the critical load exceedances and the related effects are indeed to a some extent observed.

Despite these effects, the Speulder forest grows well and can be classified as moderately vital, using needle loss and needle discoloration as vitality parameters (Steingröver and Jans, 1994). Compared to other Dutch forests in the Netherlands, the vitality of the trees at the Speulder forest decreased from 1986 to 1994. Besides the postponed thinning, the gradual decline of the nutrient status of the trees may be the cause for this. The N content in the needles is at its optimum for biomass production, but indications of excess N were found: poor mycorrhization, high foliage/fine root ratios and high arginine contents in the needles. The foliage/fine root ratio and arginine content exceeded the values found in clean areas by a factor 2 and 5-10, respectively.

No relation was found between the vitality class of an individual tree and its total needle surface area (Steingröver and Jans, 1995), suggesting that, at least at the Speulder forest, needle loss is not a good indicator of tree vitality. There is a natural variation in tree vitality in monoculture forests as a result of local stand conditions (soil, soil water potential, mychorriza, etc.) and genetic differences. Local stand conditions are very variable at Speulder forest (see section 7.3.4). The ecological principle of competition assumes that all organisms in a forest differ genetically in resistance and susceptibility to all types of stress (Woodman and Cowling, 1987). This natural variation in forest condition makes it difficult to detect additional changes induced, for example, by airborne chemicals.

Reference situation

Effects may also be hard to detect because of a lack of a reference: how would the Speulder forest look if it had not been exposed to the levels of anthropogenic pollution, as it has been during the past decades, and/or when thinning had not been postponed. A clue for a reference may be found in old statistics of the forestry service, which were used to estimate wood production expectations. The Speulder forest can be classified as fast growing according to the predictions of yields for production forests in the Netherlands, based on growth data recorded in the fifties (LaBastide and Faber, 1972). This is not expected since the stand was not thinned in the last decade and may be an indication that high nitrogen loads lead to higher growth rates. A negative effect of nitrogen availability and growth stimulation is the increase in the foliage/fine root ratio. At the Speulder forest, this ratio is high compared to similar Douglas fir stands in non-polluted areas. In the future the N utilisation efficiency will probably decrease and the primary production will be limited by other essential resources than nitrogen. This is shown by the decreasing K^+ concentration and increasing N/K^+ ratio in the needles from 1987 onwards, the already existing K^+ and P deficiency, the deficient N/P and N/Mg^{2+} ratio and the arginine accumulation in the needles. The relative low root/mycorrhiza density will reinforce the effect of drought and/or nutrient deficiency as a result of increased N availability and/or soil acidification (Al^{3+} toxification). Dry years or a sequence of dry years may enhance the effects of air pollutants and soil acidification on tree health. Furthermore, a relatively low root density will increase the risk of windthrow.

Manipulation experiments

Another method to introduce a reference situation in some way is by using the results of the manipulation experiments. Within the framework of the NITREX programme, manipulation experiments have been conducted in the Speulder forest by covering an area under the Douglas canopy by a roof and applying pre-industrialised and collected rainwater under the roof to the soil (Boxman *et al.*, 1995a,b). These experiments are focused on recovering the quality of the soil, needles and vitality of the trees by reducing soil loads to pre-industrialised levels. Unfortunately, the exposure of the canopy to air pollutants cannot be reduced. The nitrogen and sulphur concentrations in the soil solution were found already decreased within several months, improving the nutrient balance considerably (Boxman *et al.*, 1995a,b). To date, the nitrogen content in the needles and the vitality of the trees have remained similar for the trees exposed to both pre-industrialised and current loads. This could be the result of the above-ground uptake of nitrogen by the trees. Above-ground N uptake is probably enhanced when the supply from the soil is lowered (Perez Soba and Van der Eerden, 1992). The arginine content of the needles was, however, significantly reduced, indicating that trees react first to the reduced nitrogen availability in the soil by producing less arginine (Boxman *et al.*, 1995b). In the more polluted southeastern part of the Netherlands at a similar manipulation plot, an even higher reduction of arginine in needles was found for these years (Boxman *et al.*, 1995b). The difference between the two sites might be the result of a difference in nitrogen exposure in recent years. In the southeastern part of the Netherlands larger ammonia emission reductions are reported than in the surroundings of the Speulder forest (Van der Hoek, 1994). NH_x deposition at the Speulder forest measured as throughfall showed only a small decrease in recent years (Van Leeuwen and Bleuten, 1994).

As a result of the reduction in nitrogen en sulphur inputs during the manipulation experiment, the species diversity of microarthropods was found to be increased. Moreover, the nutrient balance in the forest has improved; this was suggested to be related to an enhanced root quality, a favourable NH_4^+/NO_3^- ratio in the soil solution and decreased leaching of base cations. The nitrogen concentrations in older needles were found to be lower than in current needles, which is the normal situation in nitrogen-limited coniferous ecosystems. This points to a re-allocation of nitrogen from the older to younger needles and is consistent with the decreasing arginine concentration in the needles (Boxman *et al.*, 1995).

N budget of the Speulder forest

In 1992, the Speulder forest experienced an atmospheric N deposition of about 55 kg ha^{-1} a^{-1}. Moreover, every year about 73 kg N ha^{-1} is released due to mineralisation of the litter layer (Heij and Schneider, 1991). Since the need for this essential nutrient is great, plants have evolved very efficient mechanisms to gather and retain it. Consequently, almost no nitrogen is present in the drainage water that leaves the rooting zone of pristine forests (Vitousek *et al.*, 1979; Grennfelt and Hultberg, 1986). Unfortunately, no information on the chemical composition of drainage water in the Netherlands is available for the time period before air pollution started to increase. However, Minderman and Leeflang (1968) showed that almost no nitrogen leached from a lysimeter planted with black pine trees in the period 1947-1961. At the moment, approximately 31 kg N ha^{-1} a^{-1} is found to leach from the rooting zone in the Speulder forest (Van der Maas and Pape, 1991), indicating nitrogen saturation (Aber *et al.*, 1989). The nitrogen uptake by the needles is probably regulated by uptake of NH_3, NH_4^+, NO_2 and HNO_2 from the atmosphere. It is estimated that 5.4 kg ha^{-1} a^{-1} of nitrogen is taken up by the canopy through this route. Total below-ground uptake of N in the Speulder forest amounts to 92 kg ha^{-1} a^{-1} (Hey and Schneider, 1991). The total amount of N stored in the needles equals 344 kg ha^{-1} (Van der Maas and Pape, 1991; Steingröver and Jans, 1995). According to data from the literature this is very high. For instance, in a Douglas fir stand with trees of equal age and heavily fertilised for nine years only 115 kg N ha^{-1} was found to be stored in the needles (Pang *et al.*, 1978). This is probably the result of the very large foliage biomass in the Speulder forest.

7.3.8 CONCLUSIONS

Gaseous deposition

Continuous measurements of u_* and H for one month show that the constant flux layer assumption for momentum and heat above the Speulder forest seems valid. The results of the deposition parameters obtained from the continuous measurements of SO_2 and NH_3 vertical gradients and meteorological parameters indicate that deposition monitoring of SO_2 and NH_3 onto forest is possible. For future monitoring activities, investigation of how to reduce random errors in individual concentration measurements is recommended. Possibilities for reducing random errors are minimising tube length, enlarging the averaging time for concentration measurements and averaging gradients for estimating deposition parameters. Deposition monitoring of NO_2 with the currently used monitors is concluded to be impossible as a result of instrumental failure

Values of R_c of SO_2 for the Douglas fir forest are generally low, differences between day and night, and wet and dry conditions have been observed. An R_c parameterisation derived for heathland vegetation yields reasonable results for forests. Further improvements of the R_c parametrisation is possible by minimising errors in measured quantities, improving the parametrisation for surface wetness, and extending the parametrisation with information on buffering capacities (of leaves or the atmosphere).

Surface exchange of ammonia over a coniferous forest has been measured continuously since November 1992. Canopy wetness has been observed to have a very strong influence on the dry deposition of ammonia: the surface resistance for a wet canopy is negligible. Over a dry canopy, the observed flux generally exceeds the estimated stomatal flux, indicating that deposition to external leaf surfaces is also important. Emission of ammonia was observed to occur during the daytime at low ambient humidity (<80%), and is thought to be related to evaporation of water films on the canopy. It is estimated that up to 20% of the deposited ammonia was re-emitted during the period November 1992-March 1993. An initial surface resistance parametrisation for surface exchange of ammonia over coniferous forest was derived. Modelled deposition velocity values have been found to compare reasonably to observed values.

Deposition of particles

The state of knowledge on particle deposition is relatively uncertain. Two contradicting opinions have been aired in the literature: 1) deposition of fine particles is negligible and 2) deposition of fine particles might contribute to a large extent to acid loads in forests. The main uncertainty is in the dry deposition velocity. The aim therefore was to use all available means and combined efforts to reduce this uncertainty to more accurately determine particle deposition to forests. It was assumed that by using the results of different kinds of measurements, in combination with a large modelling effort, at least a choice could be made between the two statements. Results would then likely point in one direction despite the large uncertainty.

The most important factors leading to uncertainty in particle deposition are the measuring errors, the uncertainty in representativeness of size distribution of the measurements, the scaling factors for accumulation experiments, and the influence of chemical conversion and humidity on particle growth. In general, the experimental and modelled results show a distinct influence of the size distribution and u_* on the dry deposition velocity of particles. They show that V_d (^{214}Pb) < V_d (SO_4) < V_d (NO_3) < V_d (base cations) < V_d (fog), in line with the size distributions. The deposition velocity of fog and base cations is proportional to u_*^2, indicating that impaction is the most important process determining V_d. Sedimentation is also important. The V_d values of other compounds are proportional to u_*^x, with $0 < x < 1$, indicating that no distinct process has precedence a mixture of processes.

Size- and u_* dependence of the dry particle deposition velocity is similar for both the measurements and the modelled results. This serves as some sort of validation of the most determining processes. The overall error in modelled V_d integrated over the size distribution representative for acidifying aerosols equals about 65%. For base cations, this error is somewhat smaller (60%) because of the contribution of the relatively well-parametrised sedimentation description. The uncertainty in model estimates is lower than or about equal to the uncertainty in measurement results, with the exception of the fog deposition measurements, which are estimated to have smaller errors (20%). The fractional bias of the means (the relative difference between the mean calculated and observed values) falls within these limits. The relatively high sensitivity of the model and an inaccuracy of the same order in measuring results mean that a perfect 1:1 correspondence between calculated and observed values cannot be expected. The Wilcoxon tests for paired samples, however, revealed no significant differences between the mean values of modelled and measured fluxes or V_d's, showing that they are in good agreement.

From the results obtained in this project the deposition of fine particles can be concluded to be an important pathway for acid input to forests. Dry deposition velocities of particles to forests and probably other rough surfaces have been confirmed to be high. Six months average V_d values for fine particles in the Speulder forest range from 1 to 2 cm s^{-1} (K^+, SO_4^{2-}, NO_3^-, and NH_4^+), with daytime velocities at 1.3 ± 1.2 cm s^{-1} and night-time velocities at 1.0 ± 1.4 cm s^{-1} (SO_4^{2-}). V_d values for coarse particles are about 5 cm s^{-1}, with daytime values of 5.1 ± 3.9 cm s^{-1} and night-time values of 4.8 ± 4.0 cm s^{-1}. In comparison, V_d values for SO_2, NH_3 and NO_2 for the same period were 1.5, 2.5 and 0.1 cm s^{-1}, respectively. This means that the deposition of aerosols to forest canopies in the Netherlands is to date underestimated by a factor of 2 to 3. For forests in Europe, this is even higher for the EMEP model results.

The answers to the three main research questions which were formulated in the introduction can be now be formulated:
1. Total deposition of SOx amounted 1185 mol ha^{-1} a^{-1} in the Speulder forest in 1993. The contribution of SO_4^{2-} aerosols to the total deposition of SOx amounted to 18%; fog deposition contributed 3% and dry deposition of SO_2, 56%. Total deposition of NHx amounted to 2865 mol ha^{-1} a^{-1}; dry NH_4 aerosols contributed 23%; fog deposition, 3% and dry deposition, 50%. Total deposition of NOy amounted to 1085 mol ha^{-1} a^{-1}; dry NO_3 aerosols contributed 38%; fog deposition, 2% and dry deposition of NOy, 33%. Aerosol input for SOx and NHx is significant and forms a major input for NO_3. These results are representative for nature conservation areas in the Netherlands which are classified as rough terrains i.e. with many, isolated trees, scattered hedges, etc. For nature conservation areas consisting mainly of low vegetation, such as large areas of heathland, the aerosol deposition is much less (about 5-10% for SO_4, 10-15% for NH_4 and 5-10% for NO_3).

2. The results of the comparison study of atmospheric deposition and throughfall fluxes, and the experiments to determine canopy exchange (rinsing of artificial and Douglas fir branches, ^{35}S experiments), show that H^+ and NH_4^+ are taken up by the canopy. Uptake of H^+ and NH_4^+ is compensated for by the leaching of Mg^{2+}, Ca^{2+}, and most of all, K. SO_2 taken up by stomata is eventually leached again, whereas NH_3 taken up via stomata is probably not leached from the canopy. Oxidised nitrogen components, especially NO_2,, are taken up by the stomata in the

canopy. Whether NO_3^- is taken up is uncertain. Na^+ and Cl^- are considered inert. The highest uncertainty is found in the estimates of the nitrogen components. The contribution of particle deposition to the throughfall flux is considerable for base cations. Along with leaching from root-derived nutrients (except for Na^+), the throughfall flux of base cations is the result of particle deposition. The deposition of acidifying aerosols to rough forests has been, to date, underestimated, thereby closing the gap between throughfall fluxes of sulphur and atmospheric deposition estimates; there is no net uptake or loss of SO_4^{2-}. Throughfall fluxes of NH_4^+ and atmospheric deposition are in reasonable agreement; aerosol fluxes of NH_4^+ appear in the order of uncertainty found in the two methods. For NO_3^-, however, the systematic difference between atmospheric deposition and throughfall fluxes has increased with the new estimates of aerosol input. There is still a large uncertainty in canopy exchange processes for oxidised nitrogen and in deposition estimates of the different gases contributing to the total oxidised nitrogen flux.

The throughfall method can be concluded to be applicable for monitoring deposition of SO_4^{2-}, Na^+ and Cl^-. Uncertainty in the results obtained with throughfall is in the same order of magnitude as that in atmospheric deposition estimates on an annual basis. The throughfall, however, has to be representatively collected for the whole canopy (e.g. by using gutters) and samples must not remain in the field longer than one week. For the Speulder forest it has been shown that throughfall measurements might also be used for Mg^{2+}, K^+ and Ca^{2+}, provided the model of Van der Maas (1990) is used to correct for canopy exchange. It is uncertain whether the model can be used for measurements in other pollution climates and/or for other tree species. Results for nitrogen components are uncertain. It seems that for conditions found in the Speulder forest, the throughfall method can also be applied for NH_4^+; this leads to estimates similar to atmospheric deposition. For NO_3^-, it is not advisable to use the throughfall method until uncertainty in canopy exchange processes is reduced.

3. Sulphur is estimated to be deposited at 1185 mol ha^{-1} a^{-1} on the canopy and the forest soil., while 3950 mol ha^{-1} a^{-1} of nitrogen is deposited onto the canopy, of which 3565 mol ha^{-1} a^{-1} reaches the soil surface. For Na^+ and Cl^-, about 1220 and 1425 mol ha^{-1} a^{-1}, respectively, deposit on the canopy and reaches the soil unchanged. For K^+, 325 mol ha^{-1} a^{-1} reaches the soil surface; 75 mol ha^{-1} a^{-1} is of atmospheric origin, whereas 250 mol ha^{-1} a^{-1} is root derived K^+. Regarding Mg^{2+}, 225 mol ha^{-1} a^{-1} reaches the soil surface: of this 185 mol ha^{-1} a^{-1} is of atmospheric origin. For Ca^{2+}, these numbers are 210 mol ha^{-1} a^{-1} and 150 mol ha^{-1} a^{-1} respectively. Total base cation input (Ca^{2+}+K^++Mg^{2+}), 390 mol ha^{-1} a^{-1}, represents about 6% of the total potential acid deposition of 6320 mol ha^{-1} a^{-1}. The base cation deposition forms therefore an important input in forests in the Netherlands.

Assessment of the relation between loads/levels and effects

The Speulder forest is a well growing forest, exceeding production forest yield predictions based on data observed in the fifties. This large growth rate is the result of high nitrogen exposure and loads during the last few decades. The high foliage/fine root and foliage/large root ratio observed is also the result of high nitrogen loads, making the forest increasingly sensitive to drought, storms, frost and parasite injury (insects, fungi, virus). Mycorrhiza are poorly developed in the Speulder forest. There are indications that this is also caused by excess nitrogen availability but more information is needed to confirm this.

The elevated arginine concentrations in the needles indicate excessive nitrogen availability as do the disturbed balances between nitrogen and other nutrients. So far, it is not known whether arginine accumulation is detrimental to trees. In large concentrations it was found to affect cellular pH regulation and may be associated with increased parasite susceptibility. P and K are deficient in the needles. Ratios of most nutrients to nitrogen indicate that the nutrient status of the trees is far from optimal and gradually getting worse. The N utilisation efficiency of the Speulder forest will decrease in the future. The primary production will become limited by other essential resources than N. Nitrate is leaching from the rooting zone in large quantities and in this respect the Speulder forest ecosystem can be regarded as nitrogen saturated. Aluminium is also leaching in large amounts.

NH_4^+ concentrations in water layers covering the needle surface were sometimes such that damage of the epicuticular wax layer can be expected. However, this effect was not investigated in the Speulder forest. Over the period of a year, CO_2 assimilation was reduced as a result of ozone exposure and high vapour pressure deficits. The *AOT40* of 10 ppm h set to protect tree species against a growth reduction of more than 10% seems too small. Exposure to high O_3 and/or high NH_3 concentrations probably cause stomata to remain open, even at low shoot-water potentials and/or high vapour pressure deficits. In this way trees become more vulnerable to drought. Short periods with considerable water stress have occurred. As high aluminium concentrations are found in these periods, for example, the combination of drought stress and stress by unfavourable soil chemical conditions may cause adverse effects.

It is difficult to draw conclusions with respect to tree or stand vitality both now and in the future, as it depends on how vitality is defined. If vitality is defined in the traditional terms of needle loss and discoloration, no adverse effects are seen at the Speulder forest. However, poorly developed mycorrhiza may be regarded as an indication of reduced vitality as this implies a reduced capacity to take up water and nutrients. Besides the nutrient status of the needles, the high foliage/fine root and foliage/coarse root ratio may be regarded to indicate future risk for vitality reduction.

Exceedance of critical levels and loads means that there is a greater risk that adverse effects occur. The risk will be higher with larger exceedances and a longer duration of the exceedance. Whether such effects occur as a result of exceedances depends on the interaction with other stresses. The Speulder forest is under stress and, for this reason, more vulnerable to other stresses. The risk that adverse effects occur, cannot be properly quantified with current knowledge. A more quantitative risk assessment for forests should be developed in the near future by using results from experiments and by modelling (Van Grinsven *et al.*, 1994).

7.3.9 EVALUATION

The aim of the combined research in the Speulder forest was to determine the exposure and loads of acidifying and eutrophying pollutants, as well as their harmful influence on the vitality of Douglas fir trees and the forest ecosystem as a whole. The question is whether this research has succeeded. The success of a research project is related to the state of previous knowledge: is the research for validating known causal relations in the field, for investigation of unknown processes or for evaluating expectations of effects by measuring all that can be measured?

When research in the Speulder forest started in 1985, the state of knowledge on acidification was such that damage to ecosystems was observed in very highly polluted areas; also, in less polluted areas damage was expected to be forthcoming. The eutrophication problem had only been just recognised. It was not known at that time which combinations of stress was causing the damage and what local conditions could prevent or reduce damage.

The Speulder forest was at the time of selection a stand showing good growth. The criteria for selection of the stand were related to species, representativeness, conditions and surroundings, but also to the possibility of measuring the deposition using micrometeorological techniques (homogeneity of the stand and its surroundings, no sources in the neighbourhood, no advection problems, etc.). This limited the choice of forest stands in the Netherlands considerably. Furthermore, it appeared impossible to fulfil all criteria (Vermetten *et al.*, 1989).

Research of this kind should focus on both the causal relations and processes which might play a role in the cause - effect chain. Processes can be studied under controlled conditions in the laboratory or in open-top chambers, where process-related parameters can be manipulated. Field studies are always necessary to confirm the laboratory studies, e.g. to see if feedback mechanisms occur. The manipulation experiment is a good example of research on such processes.

Deriving mono-causal relations from the multiple stress to which the Speulder forests is exposed is difficult because of a lack of a proper reference and/or a lack of results from comparable investigations allowing comparison of temporal and spatial variations in (changes) in different effect parameters. This means that this kind of research should be carried out at more locations under different environmental circumstances as a combined effort of institutes and disciplinary-oriented bodies. Furthermore, as the observed effects are the result of multiple stress, all stress parameters should be investigated and not only those related to acidification, eutrophication and ozone.

The resilience of forests is high: relatively old ecosystems have large enough buffering capacities to cope with multiple stresses for several years. In an effort to determine effects of environmental stress, it is therefore advisable to continue research for several years.

Monitoring programmes should be established for effect and stress parameters, and process level investigations have to be repeated frequently during the monitoring programmes. Simultaneous research at different locations under different environmental conditions would enhance the chance of deriving mono-causal relations.

Manipulation experiments can serve in the introduction of a reference situation, so as to observe the difference between a polluted and non-polluted situation. The above-ground exposure, which is not reduced using a roof, presents a problem. The NITREX experiments were too short to see what the long-term recovery of the trees would be, or what the 'normal' growth (reference situation) would be. Extending the manipulation experiment at the Speulder forest site is therefore desirable. Another way of introducing a reference is using forest production yield predictions based on observations in the fifties.

Last but not least, in such large research programmes, where the experimental effort is enormous, more time should be reserved for interpretation of the large amount of experimental data.

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