

# Chapter 15

## Atmospheric Deposition and Canopy Interactions

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### 15.1 Introduction

Element inputs by atmospheric deposition form a major contribution to a number of element fluxes of forest ecosystems. During the last few decades, inputs from the atmosphere have significantly altered the geochemical cycles of forest ecosystems especially in heavily polluted areas of Central Europe where forests have remained major sinks for air pollution.

The deposition of acids such as sulphuric and nitric acids was a major environmental concern during recent decades (Galloway 1995). Acid deposition has caused the acidification of soils and freshwaters in large areas of North America and Europe (Johnson et al. 1991). The effects of soil acidification are the leaching of base cations from the soil (Matzner and Murach 1995), the release of aluminium (Al) ions and heavy metals into soil solution (Tyler 1983), reduced decomposition of soil organic matter, and reduced growth of fine roots (Godbold et al. 2003). The release of acid soil solutions to the hydrosphere is detrimental to aquatic ecosystems.

During the last two decades, deposition of acids has decreased substantially in Central Europe due to improved emission controls and the closing down of industry in eastern Germany after the reunion (Meesenburg et al. 1995). However, the deposition of nitrogen (N) compounds has decreased only slightly and has become an increasingly important fraction of the total deposition of acids (Wright et al. 1995). Despite reduced deposition of free acidity, the acid load to soils is still high because of high deposition of ammonium ( $\text{NH}_4^+$ ). The fate of elevated N-input on forest ecosystem remains partly unknown, but besides the impact on the acid/base balance of ecosystems there are some indications of increased tree growth, reduced root/shoot ratio, nutrient imbalances, reduced frost hardiness and elevated foliage consumption by insects (Binkley and Högberg 1997; Aber et al. 1998; Meiwes et al. 1999). In N-saturated ecosystems, soils have increased susceptibility for losses of nitrate ( $\text{NO}_3^-$ ) to the hydrosphere and of trace gases to the atmosphere (Aber 2002).

The transfer of elements from the atmosphere to forests takes several pathways. According to Ulrich (1994), total deposition can be divided into wet deposition and

interception deposition. The latter is composed of particulate interception and gaseous interception and is also referred as dry deposition. After deposition to the canopy of forest stands, the deposited elements may either be taken up by the canopy or washed from the surfaces by subsequent rainfall (Harrison et al. 2000).

Precipitation beneath the canopy of forest ecosystems contains both wet deposition and interception deposition. As the canopy acts as sink or source for solutes in precipitation passing through the canopy, stand precipitation cannot be used as a measure of total deposition (Horn et al. 1989). Total deposition can be calculated using wet deposition and independent estimates of dry deposition. Frequently used methods for the estimation of dry deposition are: canopy budget models (Draaijers et al. 1996), inferential modelling (van Leeuwen et al. 1996; Gauger et al. 2002) and gradient measurements of air pollutants (Sutton et al. 1995). The use of canopy budget models for forest ecosystems was extensively discussed by Draaijers (1999). Results of canopy budget models are very uncertain for the estimation of N-deposition, because dry deposition can form a high proportion of total deposition of N (Lindberg et al. 1986), and N is involved intensively in interaction processes with the foliage (Horn et al. 1989).

For the investigation of effects of atmospheric deposition on nutrient cycles of forest ecosystems and of temporal trends of element fluxes on ecosystem processes, long-term monitoring sites are of overwhelming importance. For this study, three mature beech forests located at the northern part of the central German mountain range have been compared. The three sites (Solling, Göttinger Wald and Zierenberg) are included in the Level II European Forest Intensive Monitoring programme (de Vries et al. 2001). At the Solling site, deposition measurements started in 1968. The Solling beech forest site together with the Solling spruce forest site has – to our knowledge – the longest continuous record of throughfall measurements globally. Deposition measurements at the Göttinger Wald site started in 1981 and at Zierenberg in 1989.

In this chapter, these long-term data sets will be used: (1) to characterise the chemical composition of open field deposition, throughfall and stemflow and to analyse relationships among major input components to relate them to different sources, (2) to describe the annual deposition fluxes and their temporal changes on the three sites, and (3) to analyse the interactions of precipitation inputs with the canopy of the stands.

*Data collection and evaluation procedures.* Element fluxes have been measured in open field deposition, throughfall and stemflow. Stand precipitation is the sum of throughfall and stemflow. Open field deposition and throughfall were monitored with samplers, which remained continuously open to the atmosphere (bulk samplers). Samples obtained with such samplers are composed of rainwater or snow and gravitational sedimented particles. In remote areas, there is little difference between element fluxes of bulk precipitation and wet-only precipitation (Ibrom 1993; Gauger et al. 2002). Open field deposition is being sampled at clearings located close to the monitoring sites. Here, we use the term open field deposition instead of bulk deposition, because the term bulk deposition is generally used for sampling of precipitation with bulk samplers without any regard to their location

(open field or under the canopy). At the Solling and Göttinger Wald sites, samplers with 50-cm<sup>2</sup> surface area were used in summer months (May–October) until 1990. From 1990 onwards, funnel-flask samplers with 87.5-cm<sup>2</sup> surface area have been used for summer sampling. In winter months (November–April), buckets with surface area of 570 (until 1990) or 500 cm<sup>2</sup> (from 1990 onwards) have been used (Meiwes et al. 1984). At the Zierenberg site, funnel-flask samplers with 100-cm<sup>2</sup> surface area have been used in summer months and buckets with 500-cm<sup>2</sup> surface area in winter months (Brechtel and Hammes 1984; Eichhorn 1995). At the Solling and Göttinger Wald sites, six samplers have been used for open field deposition and 15 samplers for throughfall (Meesenburg et al. 1997). At the Zierenberg site, ten replicates have been used for open field deposition and 20 for throughfall. Coarse particles (e.g. litter) have been prevented from falling into the samplers by using a polyethylene mesh at the Solling and Göttinger Wald sites and a ceramic sieve at Zierenberg site. After recording the volumes, three composite samples were formed for each of open field deposition and throughfall for the Solling and Göttinger Wald sites, and four composite samples for the Zierenberg site for laboratory analysis.

Sampling devices and sampling procedures of the three sites were tested in comparison with 18 other methods for open field deposition and throughfall used within the framework of the ICP forest level II programme. The performance of the deposition monitoring at the study sites was found to be acceptable (Draaijers et al. 2001).

Stemflow has been sampled by fixing polyurethane spirals around the stems, which were coated with paraffine. Three to five replicates were installed, which were analysed separately (Solling and Göttinger Wald sites) or pooled to a composite sample (Zierenberg site).

Water flux via stemflow for Zierenberg and Solling has been estimated to be 15% of the total throughfall flux. At the Solling site, a value of 15% is close to the mean value of stemflow flux estimated by Benecke (1984) for the period 1969–1975. For Göttinger Wald, stemflow fluxes were obtained from regression functions between stemflow volume and throughfall (Gerke 1987).

Analytical methods are described by Fassbender and Ahrens (1977) and König and Fortmann (1996a–d) for the Solling and Göttinger Wald sites. In short, pH was measured potentiometrically. Sodium (Na<sup>+</sup>), potassium (K<sup>+</sup>), magnesium (Mg<sup>2+</sup>), calcium (Ca<sup>2+</sup>), and manganese (Mn<sup>2+</sup>) were determined by AAS until 1989, and since 1990 by ICP-AES. NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> and chloride (Cl<sup>-</sup>) were analysed colorimetrically with a continuous flow system. Sulphate (SO<sub>4</sub><sup>2-</sup>) was measured by precipitation with Ba and by potentiometric titration of excess Ba with EDTA until 1982, from 1983 to 1992 by the methyl-thymol-blue method, and since 1993 by ICP-AES. Organic N (N<sub>org</sub>) is calculated as the difference between total N (N<sub>tot</sub>) (measured after digestion) and the sum of NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup>.

Because no independent estimates of interception deposition are available for the study sites over the whole observation period, we have used the canopy budget model developed by Ulrich (1994) for the calculation of total deposition. Annual fluxes of ions have been used for the calculation procedure. The canopy budget model of Ulrich (1994) estimates the interception deposition for element species *A* (ID<sub>A</sub>), which are not adsorbed or leached from the canopy, from the difference of

stand precipitation ( $SP_A$ ) and open field precipitation (wet deposition  $OF_A$ ). Interception deposition  $ID_A$  is the sum of particulate ( $ID_{part,A}$ ) and gaseous deposition ( $ID_{gas,A}$ ). Total deposition ( $TD_A$ ) is the sum of wet deposition and interception deposition.

$$TD_A = OF_A + ID_A, \quad (15.1)$$

$$ID_A = ID_{part,A} + ID_{gas,A}, \quad (15.2)$$

$$ID_A = SP_A - OF_A. \quad (15.3)$$

$$A = \text{Na, Cl, SO}_4.$$

Particulate interception deposition ( $ID_{part,A}$ ) is estimated from the ratio between interception deposition and open field precipitation of Na assuming that  $\text{Na}^+$  is only deposited by wet deposition and particulate interception. It is assumed that the particle size distribution of all deposited substances is similar, resulting in a similar deposition velocity. Another assumption is that particulate interception is caused to a large degree by fog droplets. In contrast to earlier formulations of the model by Ulrich (1983) and Bredemeier (1988), Ulrich (1994) extended the model for  $\text{NH}_4^+$  and  $\text{NO}_3^-$  implying the assumption of similar deposition velocities holds for particulates containing  $\text{NH}_4^+$  and  $\text{NO}_3^-$  particles. The assumption of similar deposition velocities is highly questionable especially for N-compounds (Spranger 1992):

$$f_{\text{Na}} = \frac{ID_{\text{Na}}}{OF_{\text{Na}}}, \quad (15.4)$$

$$ID_{part,A} = f_{\text{Na}} OF_A. \quad (15.5)$$

$$A = \text{H, K, Mg, Ca, Mn, Al, Fe, NH}_4, \text{Cl, SO}_4, \text{NO}_3.$$

Gaseous deposition ( $ID_{gas,A}$ ) of metal cations is assumed to be negligible.

$$ID_{gas,A} = 0. \quad (15.6)$$

$$A = \text{Na, K, Mg, Ca, Mn, Al, Fe.}$$

Gaseous deposition ( $ID_{gas,A}$ ) of  $\text{SO}_2$ ,  $\text{HCl}$ ,  $\text{HNO}_3$  and  $\text{NH}_3$  is estimated from the difference of interception deposition and particulate deposition:

$$ID_{gas,A} = ID_A - ID_{part,A} = SP_A - OF_A - ID_{part,A}. \quad (15.7)$$

$$A = \text{NH}_4, \text{Cl, SO}_4, \text{NO}_3.$$

Gaseous deposition of  $\text{SO}_2$ ,  $\text{HCl}$  and  $\text{HNO}_3$  causes an equivalent input of protons, gaseous deposition of  $\text{NH}_4^+$  a consumption of protons:

$$\text{ID}_{\text{gas,H}} = \text{ID}_{\text{gas,SO}_2} + \text{ID}_{\text{gas,Cl}} + \text{ID}_{\text{gas,NO}_3} - \text{ID}_{\text{gas,NH}_4}. \quad (15.8)$$

If particulate deposition of  $\text{NH}_4^+$  and  $\text{NO}_3^-$  is higher than the difference between  $\text{SP}_A$  and  $\text{OF}_A$ , no gaseous deposition can be calculated.

The difference between total deposition and stand precipitation is interpreted as canopy budget ( $\text{CB}_A$ ). Positive values are interpreted as leaching, negative values as uptake by the canopy:

$$\text{CB}_A = \text{SP}_A - \text{TD}_A = \text{SP}_A - \text{OF}_A - \text{ID}_A. \quad (15.9)$$

From the calculation scheme, it arises that either gaseous deposition (if  $\text{ID}_{\text{part}} < \text{SP}_B - \text{OF}_B$ ) or uptake by the canopy (if  $\text{ID}_{\text{part}} > \text{SP}_B - \text{OF}_B$ ) is calculated for  $\text{NH}_4^+$  and  $\text{NO}_3^-$ . As both processes can occur concurrently (Veithen 1996; Garten et al. 1998), total deposition of  $\text{NH}_4^+$  and  $\text{NO}_3^-$  is underestimated by the model. Moreover, the processes of canopy uptake and leaching are highly seasonal and the use of annual budgets disregards the seasonal nature of these processes. The assumptions involved in the model were not tested on these three sites and will need due consideration during the interpretations of model results.

Despite the limitations mentioned above, the canopy model of Ulrich (1994) has been applied to  $\text{NH}_4^+$  and  $\text{NO}_3^-$ . Total deposition of nitrogen ( $\text{N}_{\text{tot}}$ ) has been calculated as the sum of total deposition of  $\text{NH}_4^+$ , total deposition of  $\text{NO}_3^-$  and open field deposition of  $\text{N}_{\text{org}}$  (Ulrich 1994).

## 15.2 Precipitation Chemistry

For characterisation of the chemical composition of atmospheric deposition, data for the period 1993–1998 were selected as all three study sites had information for this period. As precipitation chemistry has changed significantly during the last few decades, only the pattern for the selected period is described.

At the Solling site, open field precipitation chemistry is dominated by  $\text{NH}_4^+$  (44% of cations on equivalent basis) and  $\text{Na}^+$  (23%), and  $\text{H}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{K}^+$  are only of minor importance in that order. Anions of significance are  $\text{SO}_4^{2-}$  (43%),  $\text{NO}_3^-$  (35%) and  $\text{Cl}^-$  (22%). Concentrations of  $\text{NH}_4^+$  are higher than concentrations of  $\text{NO}_3^-$  (Table 15.1). The contribution of  $\text{N}_{\text{org}}$  to  $\text{N}_{\text{tot}}$  is about 7%.

At the Göttinger Wald site, 44% of the sum of cations in open field precipitation is  $\text{NH}_4^+$ .  $\text{Na}^+$  (19%),  $\text{Ca}^{2+}$  (15%), and  $\text{H}^+$  (10%) also have some quantitative importance. The contribution to the sum of anions is 16% for  $\text{Cl}^-$ , 39% for  $\text{NO}_3^-$  and 44% for  $\text{SO}_4^{2-}$ .

At the Zierenberg site,  $\text{NH}_4^+$  (30%) is the most abundant cation in open field precipitation, but the relative contributions of  $\text{Ca}^{2+}$  (27%) and  $\text{Mg}^{2+}$  (14%) are

**Table 15.1** Mean concentrations and standard deviation (in parentheses) of total N ( $N_{\text{tot}}$ ) and relative contribution of  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and  $N_{\text{org}}$  to  $N_{\text{tot}}$  in open field precipitation, throughfall and stemflow from 1993 to 1998 at the study sites

Site		$N_{\text{tot}}$ ( $\text{mmol l}^{-1}$ )	$\text{NH}_4^+$ %	$\text{NO}_3^-$ %	$N_{\text{org}}$ %
Solling	Open field deposition	125 (1)	52	41	7
	Throughfall	260 (18)	51	37	12
	Stemflow	216 (28)	41	40	19
Göttinger Wald	Open field deposition	170 (1)	50	43	7
	Throughfall	293 (5)	49	40	11
	Stemflow	303 (43)	44	42	15
Zierenberg	Open field deposition	174	50	41	9
	Throughfall	454	39	51	10
	Stemflow	310	45	35	20

much higher than at the Solling and Göttinger Wald sites. This may be related to higher input of dust particles and to other factors relating to methodology of collection and analysis. The contribution of  $\text{Na}^+$  to the sum of cations is 16%. The most abundant anion is  $\text{SO}_4^{2-}$  (45%), which is followed by  $\text{NO}_3^-$  (34%) and  $\text{Cl}^-$  (21%). Various anions have similar fractions in the open field precipitation on the three sites with values following the order:  $\text{SO}_4^{2-}$  (43–45%) >  $\text{NO}_3^-$  (34–39%) >  $\text{Cl}^-$  (16–22%).

$N_{\text{tot}}$  concentrations are generally highest in throughfall and lowest in open field precipitation (Table 15.1). After passing through the canopy, concentrations of  $N_{\text{tot}}$  in the precipitation increased by 108% at Solling, 72% at Göttinger Wald and 161% at Zierenberg compared to open field precipitation. This indicates a much higher interception of dry deposition at Zierenberg. The increase of concentrations of  $N_{\text{tot}}$  in stemflow was very similar at the three study sites (73–78%).

The relative contribution of the N species to  $N_{\text{tot}}$  is similar at the three study sites (Table 15.1).  $\text{NH}_4^+$  concentrations in open field deposition are higher than  $\text{NO}_3^-$  at all sites indicating that N-inputs are influenced to a large degree by animal husbandry. In throughfall and stemflow, the relative contribution of  $N_{\text{org}}$  to  $N_{\text{tot}}$  is generally higher than in open field precipitation indicating leaching of  $N_{\text{org}}$  from leaves and bark of the trees or from other sources of  $N_{\text{org}}$  in the canopy. At the Zierenberg site, the enrichment of the N-compounds in throughfall as compared to open field precipitation is much higher than at the Solling and Göttinger Wald sites. The enrichment of  $\text{NH}_4^+$  in throughfall is higher than that of  $\text{NO}_3^-$  at the Solling and Göttinger Wald sites and lower at the Zierenberg site. The strong enrichment of  $\text{NO}_3^-$  relative to  $\text{NH}_4^+$  in throughfall at Zierenberg may be explained by nitrification of  $\text{NH}_4^+$  in the canopy (Papen et al. 2002). (Table 15.1). However, Eichhorn (1995) attributed the enrichment of  $\text{NO}_3^-$  in throughfall to its leaching from the canopy. At the Solling and Göttinger Wald sites, the relative contribution of  $\text{NH}_4^+$  in stemflow is lower than in throughfall. This pattern may be partly explained by nitrification of  $\text{NH}_4^+$  at branches and stems.

Mean pH in throughfall is higher than in open field deposition at the Göttinger Wald and Zierenberg sites reflecting the buffering of acids in the canopy, but lower at Solling. pH is generally lower in stemflow than in open field deposition.

Sea spray is a major source for  $\text{Cl}^-$ ,  $\text{Na}^+$ , and  $\text{Mg}^{2+}$  in open field precipitation. An influence of road salt can be excluded since the sampling sites are far away from roads. Higher concentrations of  $\text{Cl}^-$  during the winter season are related to more frequent storm events. At the Solling site, the contribution of sea spray is 100% for  $\text{Cl}^-$ , 77% for  $\text{Na}^+$  and 44% for  $\text{Mg}^{2+}$  (calculated with  $\text{Cl}^-$  as index element). At the Göttinger Wald site, the influence of sea spray is somewhat lower than at the Solling site with values of 100% for  $\text{Cl}^-$ , 72% for  $\text{Na}^+$  and 29% for  $\text{Mg}^{2+}$ . Sea spray is an important source at the Zierenberg site for  $\text{Cl}^-$  (100%) and  $\text{Na}^+$  (80%) whereas a low value for  $\text{Mg}^{2+}$  (10%) points to sources other than sea spray being more important, e.g. soil dust due to agricultural activities or Mg containing particles from different industrial processes such as coal burning or handling of bulk cargo.

The covariance analysis of the concentrations of solutes in open field precipitation, throughfall and stemflow at the three sites was undertaken by employing principal component analysis (SPSS version 6.1.2). Principal component analysis was used to find the least linear combinations of the parameters which were required to explain as much of the total variance of the data as possible. The major ions  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{H}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ ,  $\text{NH}_4^+$  and  $\text{NO}_3^-$  and  $\text{N}_{\text{org}}$  were included for the analyses. Principal component analysis has been frequently used for assigning different sources to various solutes in the precipitation (Gorham et al. 1984; Feger 1986) and to describe the predominant processes occurring in ecosystems (Christophersen and Hooper 1992). Varimax rotation has been performed to find out the contribution of different processes.

As an example, results for open field precipitation at Solling are given in Table 15.2 for principal components with eigenvalues  $>1.0$ . Three components could be differentiated. Component 1, which explains 46% of the variance, has high loadings of  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$ . These ions are negatively correlated to the amount of precipitation.  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  can be ascribed to the emission of

**Table 15.2** Matrix of factor loadings (varimax rotation) and communalities of principal component analysis for concentrations of ions in open field deposition at Solling. Factor loadings above 0.6 are given in bold

	Component 1	Component 2	Component 3	Communality
$\text{Na}^+$	0.13	<b>0.97</b>	0.02	0.96
$\text{K}^+$	0.48	-0.15	<b>0.64</b>	0.67
$\text{Mg}^{2+}$	0.38	<b>0.84</b>	0.20	0.89
$\text{Ca}^{2+}$	<b>0.75</b>	0.27	<b>0.27</b>	0.71
$\text{H}^+$	0.21	-0.25	<b>-0.71</b>	0.62
$\text{SO}_4^{2-}$	<b>0.94</b>	0.21	0.08	0.93
$\text{Cl}^-$	0.09	<b>0.96</b>	-0.10	0.93
$\text{NH}_4^+$	<b>0.90</b>	0.14	0.10	0.84
$\text{NO}_3^-$	<b>0.93</b>	0.09	0.02	0.93
$\text{N}_{\text{org}}$	0.29	-0.08	<b>0.79</b>	0.72
Explained variance (%)	46.0	22.4	13.7	82.0

NH<sub>3</sub>, NO<sub>x</sub> and SO<sub>2</sub>. NH<sub>3</sub> emissions can be attributed mainly to intensive farming practices such as animal husbandry and field application of faecal materials. NO<sub>x</sub> is emitted to a high degree by vehicles, whereas SO<sub>2</sub> emissions can be ascribed mainly to large power plants. Ca<sup>2+</sup> and K<sup>+</sup> can be attributed partly to the emission of dust. Thus, component 1 describes components of air pollution in open field precipitation.

Component 2 explains about one-quarter of the variance and has high loadings of Cl<sup>-</sup>, Na<sup>+</sup>, and Mg<sup>2+</sup>. This component can be ascribed to the influence of sea spray. Cl<sup>-</sup>, Na<sup>+</sup> and Mg<sup>2+</sup> are independent of the amount of precipitation. The third component explains 14% of the variance and has high loadings of N<sub>org</sub> and K<sup>+</sup> and a high negative loading of H<sup>+</sup>, which can be ascribed to plant-based organic substances.

The first principal component of stemflow at Solling explains over 60% of the variance, and has high loadings of Mg<sup>2+</sup>, Ca<sup>2+</sup>, H<sup>+</sup> and SO<sub>4</sub><sup>2-</sup> and considerable loadings of K<sup>+</sup> and NO<sub>3</sub><sup>-</sup> (Table 15.3). Mg<sup>2+</sup>, Ca<sup>2+</sup> and K<sup>+</sup> ions are leached from the vegetation, when buffering of H<sup>+</sup> occurs whereas SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> are involved in the charge balance. Thus, the first component can be interpreted as describing the input of acid depositions and their subsequent buffering by ion exchange. The second component in Table 15.3 explains 18% of the variance and has high loadings of NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> and N<sub>org</sub> which may account for the transformation of N-species and their interaction with the different tree compartments and canopy epiphytes. The third component explains 10% of the variance and has high loadings of Na<sup>+</sup> and Cl<sup>-</sup>, which can be interpreted as sea spray.

The principal component analysis for open field precipitation at the Göttinger Wald and Zierenberg sites gives similar results as for the Solling site (Table 15.4). Also for throughfall data at Solling, principal components provided similar interpretation of results. For throughfall data at the Göttinger Wald and Zierenberg sites, canopy interactions are a major source affecting the variance of the data (Table 15.4).

For stemflow data at the Göttinger Wald and Zierenberg sites similar results as for the Solling site have been obtained (Table 15.4). In contrast to open field deposition, where the components can be interpreted as different sources of the

**Table 15.3** Matrix of factor loadings (varimax rotation) and communalities of principal component analysis for concentrations of ions in stemflow at Solling. Factor loadings above 0.61 are given in bold

	Component 1	Component 2	Component 3	Communality
Na <sup>+</sup>	0.34	0.10	<b>0.93</b>	0.99
K <sup>+</sup>	0.49	0.53	0.10	0.52
Mg <sup>2+</sup>	<b>0.83</b>	0.19	0.49	0.96
Ca <sup>2+</sup>	<b>0.92</b>	0.20	0.26	0.96
H <sup>+</sup>	<b>0.92</b>	0.20	0.26	0.93
SO <sub>4</sub> <sup>2-</sup>	<b>0.81</b>	0.50	0.26	0.97
Cl <sup>-</sup>	0.30	0.17	<b>0.92</b>	0.97
NH <sub>4</sub> <sup>+</sup>	0.07	<b>0.93</b>	0.19	0.90
NO <sub>3</sub> <sup>-</sup>	0.48	<b>0.76</b>	0.29	0.91
N <sub>org</sub>	0.13	<b>0.93</b>	-0.03	0.72
Explained variance (%)	62.4	18.1	9.6	90.1



**Table 15.4** Explained variance of principal components and sum of explained variance of principal component analysis (varimax rotation) for concentrations of ions in open field deposition (OF), throughfall (TF) and stemflow (SF) at Solling, Göttinger Wald and Zierenberg (interpretation of principal components is given by letters; the same interpretation means that the same elements have high loadings in certain components)

Site	Flux	Component 1 (%)	Component 2 (%)	Component 3 (%)	Σexplained variance (%)
Solling	OF	48.9 <sup>a</sup>	24.2 <sup>b</sup>	12.2 <sup>c</sup>	85.3
	TF	50.4 <sup>a</sup>	17.7 <sup>b</sup>	12.3 <sup>c</sup>	80.4
	SF	62.4 <sup>d</sup>	18.1 <sup>e</sup>	9.6 <sup>b</sup>	90.1
Göttinger Wald	OF	53.3 <sup>a</sup>	16.5 <sup>b</sup>	13.2 <sup>c</sup>	83.0
	TF	44.7 <sup>d</sup>	21.1 <sup>a</sup>	16.5 <sup>b</sup>	82.4
	SF	52.8 <sup>d</sup>	19.5 <sup>e</sup>	16.1 <sup>b</sup>	88.4
Zierenberg	OF	56.2 <sup>a</sup>	13.1 <sup>b</sup>	9.8 <sup>c</sup>	79.2
	TF	59.6 <sup>ba</sup>	16.4 <sup>d</sup>	8.3 <sup>f</sup>	84.3
	SF	57.7 <sup>d</sup>	17.7 <sup>e</sup>	10.0 <sup>b</sup>	85.5

<sup>a</sup>Air pollution

<sup>b</sup>Sea spray

<sup>c</sup>Organic deposition

<sup>d</sup>Canopy leaching

<sup>e</sup>N mineralisation processes

<sup>f</sup>H<sup>+</sup> buffering

solutes, transformation and interaction processes of the solutes with the vegetation seems to play an important role for the variance of the stemflow data.

### 15.3 Element Fluxes

We present the element fluxes with open field deposition, throughfall, stemflow, stand precipitation and total deposition separately for three periods of roughly a decade each (Tables 15.5–15.7). The period from 1969 to 1980 is only available for the Solling site. In 1981, the Göttinger Wald site was established and represents the beginning of the second period from 1981 to 1989. The Zierenberg site was established in 1990, which is the beginning of the third period from 1990 to 2002 and represents the period after emission control in Germany. Comparing the period 1990–2002 between the sites indicates higher fluxes of SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup> and Na<sup>+</sup> at the Solling site than at the Göttinger Wald and Zierenberg sites due to higher precipitation rates at the Solling site. N-fluxes in open field deposition were lowest at the Zierenberg site and highest at the Solling site, whereas they were quite similar in stand precipitation and total deposition at the three study sites. However, fluxes of K<sup>+</sup>, Mg<sup>2+</sup> and Ca<sup>2+</sup> are highest at the Zierenberg site.

Fluxes of most elements have decreased during the last 22 years at the Solling and Göttinger Wald sites (Tables 15.5 and 15.6). Decreasing trends have been detected for open field deposition, throughfall, stemflow and stand precipitation as

**Table 15.5** Mean annual rates of open field deposition (OF), throughfall (TF), stemflow (SF), stand precipitation (SP), and total deposition (TD) at the Solling site during the periods 1969–1980 (1971–1980 for  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{N}_{\text{tot}}$ ,  $\text{N}_{\text{org}}$ ), 1981–1989, and 1990–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Flux	Period	$\text{H}_2\text{O}$ (mm)	$\text{Na}^+$	$\text{K}^+$	$\text{Mg}^{2+}$	$\text{Ca}^{2+}$	$\text{H}^+$	$\text{Mn}^{2+}$	$\text{SO}_4^{2-}$	$\text{Cl}^-$	$\text{NH}_4^+$	$\text{NO}_3^-$	$\text{N}_{\text{tot}}$ (mmol $\text{m}^{-2}$ per year)	$\text{N}_{\text{org}}$ (mmol $\text{m}^{-2}$ per year)
OF	69–80	985	33	9	15	51	80	1.3	148	48	84	58	175	34
	81–89	1,145	36	9	11	36	78	1.4	123	47	85	69	181	28
	90–02	1,184	40	5	7	16	17	0.5	59	33	63	51	126	12
TF	69–80	729	45	48	26	96	88	10.7	207	67	75	63	201	61
	81–89	768	46	50	23	77	57	10.0	183	66	99	80	202	27
	90–02	812	46	49	14	30	13	4.0	82	48	74	59	157	24
SF	69–80	121	16	23	8	28	51	3.4	115	25	13	10	52	25
	81–89	122	16	20	7	24	44	2.8	82	22	15	15	42	16
	90–02	119	10	10	2	5	8	0.5	20	11	8	8	20	5
SP	69–80	850	61	72	33	124	138	14.1	322	92	87	73	253	86
	81–89	890	61	70	30	101	101	12.8	264	88	114	95	244	42
	90–02	931	55	59	16	35	21	4.5	102	59	82	67	178	29
TD	69–80	850	61	17	29	95	199	2.4	322	92	157	109	300	64
	81–89	890	61	15	19	62	204	2.5	264	88	143	117	289	47
	90–02	931	55	8	10	22	55	0.8	102	59	94	73	179	21

**Table 15.6** Mean annual rates of open field deposition (OF), throughfall (TF), stemflow (SF), stand precipitation (SP) and total deposition (TD) at the Göttinger Wald site during the periods 1981–1989 and 1990–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Flux	Period	H <sub>2</sub> O (mm)	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>	N <sub>tot</sub> (mmol m <sup>-2</sup> per year)	N <sub>org</sub> (mmol m <sup>-2</sup> per year)
OF	81–89	750	28	9	12	56	40	0.5	108	32	61	61	141	22
	90–02	684	24	3	7	19	11	0.2	48	20	47	45	100	8
TF	81–89	516	25	38	21	88	16	1.2	128	39	63	57	137	21
	90–02	519	28	42	13	37	5	0.5	59	32	59	50	126	17
SF	81–89	119	11	24	7	32	28	0.4	82	19	21	22	50	8
	90–02	97	7	14	2	7	3	0.1	20	8	10	11	25	4
SP	81–89	636	36	62	28	121	44	1.6	210	58	84	79	187	28
	90–02	615	35	56	15	44	9	0.6	79	41	70	61	151	22
TD	81–89	636	36	10	16	75	132	0.7	210	58	89	84	187	31
	90–02	615	35	5	9	27	37	0.4	79	41	75	68	151	13

**Table 15.7** Mean annual rates of open field deposition (OF), throughfall (TF), stemflow (SF), stand precipitation (SP) and total deposition (TD) at the Zierenberg site during the period 1990–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Flux	Period	H <sub>2</sub> O (mm)	(mmol <sub>c</sub> m <sup>-2</sup> per year)										N <sub>tot</sub> (mmol m <sup>-2</sup> per year)	N <sub>org</sub> (mmol m <sup>-2</sup> per year)
			Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>		
OF	90-02	752	26	7	19	38	18	0.2	45	22	39	36	85	9
TF	90-02	527	46	67	44	72	8	0.5	75	54	54	69	143	21
SF	90-02	77	7	13	5	9	3	0.1	16	9	8	7	20	5
SP	90-02	604	53	80	49	82	11	0.5	91	63	62	76	164	25
TD	90-02	604	53	14	39	77	61	0.4	91	63	81	76	171	19

well as for total deposition. Significant trends can be observed for  $\text{SO}_4^{2-}$ ,  $\text{H}^+$ ,  $\text{Cl}^-$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mn}^{2+}$  and N-species (Table 15.8). In open field deposition, stemflow and total deposition, decreasing trends are also visible for  $\text{K}^+$ . Fluxes of  $\text{SO}_4^{2-}$  decreased by 81–94% during the last 22 years and reflect the general trend of S emissions in Germany (Fig. 15.1) (Umweltbundesamt 2000; Gauger et al. 2002). Decreasing deposition rates of  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  can be attributed to reductions of dust emission in Central Europe. At the Zierenberg site, similar trends were observed although due to a shorter observation period the relative reduction of deposition was less than for the Solling and Göttinger Wald sites (Table 15.7). Significant negative trends can be observed for  $\text{SO}_4^{2-}$ ,  $\text{H}^+$ ,  $\text{Ca}^{2+}$  and  $\text{Mn}^{2+}$  at Solling even though water fluxes increased during the period 1990–2002. For  $\text{Cl}^-$ , increasing fluxes have been measured under the canopy at the Zierenberg site. Decreasing deposition rates for many elements have been observed also for other forest ecosystems in Lower Saxony (Meesenburg et al. 1995) and Hesse (Balazs 1998) as well as for most parts of Germany (Gauger et al. 2002; Matzner et al. 2004).

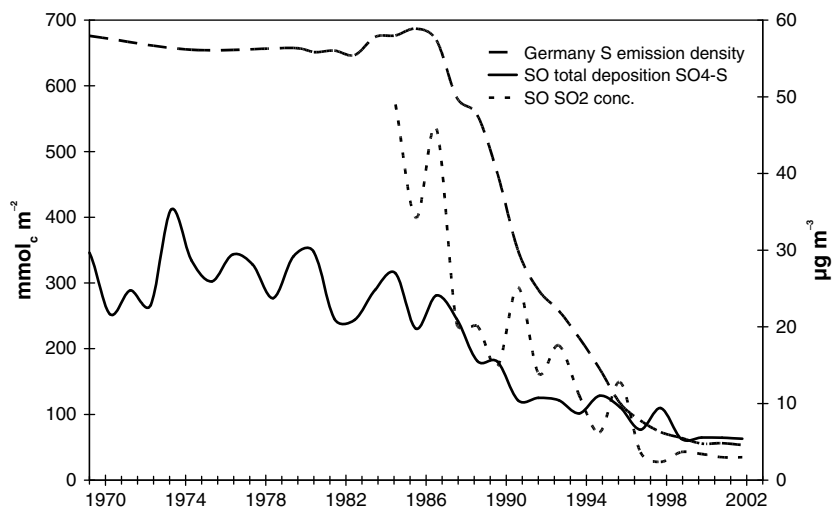
N deposition at the Solling site showed different trends during the periods 1971–1985 and 1985–2002. During the first period, fluxes of  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and  $\text{N}_{\text{tot}}$  showed no change or slightly increasing trend (Fig. 15.2). However, since 1985, a slightly decreasing trend was evident.  $\text{N}_{\text{tot}}$  fluxes have reduced by 30–55% during the period of 1981–2002 from about 36 to 22  $\text{kg ha}^{-1}$  per year. Fluxes of  $\text{NH}_4^+$  and  $\text{NO}_3^-$  with stemflow decreased even more. For  $\text{N}_{\text{org}}$ , high flux rates occurred in the 1970s, whereas low rates were measured afterwards. Temporal trends of  $\text{N}_{\text{org}}$  fluxes should be treated cautiously, since  $\text{N}_{\text{org}}$  is calculated as the difference between  $\text{N}_{\text{tot}}$  and the inorganic N-components and any alteration in the analytical methods of any one of the three components may have substantially affected the  $\text{N}_{\text{org}}$  estimates.

A decrease in N-fluxes was observed at Göttinger Wald during the whole observation period (Table 15.8, Fig. 15.3). However, for  $\text{NH}_4^+$ ,  $\text{N}_{\text{org}}$ , and  $\text{N}_{\text{tot}}$  in throughfall, these trends were not significant.  $\text{NH}_4^+$  fluxes at the different pathways decreased by 30–50% within the last two decades. Reduction of  $\text{NO}_3^-$  was about 40% for stand precipitation.  $\text{N}_{\text{tot}}$  fluxes have reduced by about 35% (from about 29–17  $\text{kg ha}^{-1}$  per year). For stemflow, relative reductions have been generally higher.

N fluxes during the period 1990–2002 were generally very similar at the study sites and the interannual variations showed the same pattern (Fig. 15.3, Tables 15.5–15.7). Open field N-deposition was highest at Solling and lowest at Zierenberg, whereas N-values in stand precipitation and total deposition were somewhat lower at Göttinger Wald than at Solling and Zierenberg. For a spruce stand at Solling, Ibrom et al. (1995) calculated by use of micro-meteorological methods a total deposition of N of 460  $\text{mmol m}^{-2}$  per year, whereas total deposition at the same stand calculated with the model of Ulrich (1994) was only 285  $\text{mmol m}^{-2}$  per year. Marques et al. (2001) showed that dry deposition (particulate and gaseous) contributed 75% to total deposition at the Solling spruce stand. These results and similar results from other locations (Harrison et al. 2000; Zimmerling et al. 2000; Meeseburg et al. 2005) suggest that the canopy model may be underestimating total deposition of N (see Horn et al. 1989; Harrison et al. 2000).

**Table 15.8** Trends of solute fluxes with open field deposition (OF), throughfall (TF), stemflow (SF), stand precipitation (SP), and total deposition (TD, according to Ulrich 1994) at the Solling and Göttinger Wald sites from 1981 to 2002 and at the Zierenberg site from 1990 to 2002 (test of trend with correlation coefficient after Pearson; o no trend, + significant increasing trend  $p \leq 0.05$ , ++ highly significant increasing trend  $p \leq 0.01$ , – significant decreasing trend  $p \leq 0.05$ , – – highly significant decreasing trend  $p \leq 0.01$ )

Site	Flux	H <sub>2</sub> O	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>	N <sub>tot</sub>	N <sub>org</sub>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>
Solling	OF	o	o	–	–	–	–	–	–	–	–	–	–	–
	TF	o	o	o	–	–	–	–	o	–	–	o	–	–
	SF	o	–	–	–	–	–	–	–	–	–	–	–	–
	SP	o	–	o	–	–	–	–	–	–	–	o	–	–
	TD	o	–	–	–	–	–	–	–	–	–	o	–	–
Göttinger Wald	OF	o	–	–	–	–	–	–	–	–	–	–	–	–
	TF	o	o	o	–	–	–	–	o	–	o	o	–	–
	SF	–	–	–	–	–	–	–	–	–	–	–	–	–
	SP	o	o	o	–	–	–	–	–	–	–	–	–	–
	TD	o	o	–	–	–	–	–	–	–	–	–	–	–
Zierenberg	OF	++	o	–	o	o	–	–	o	o	o	o	–	o
	TF	++	o	o	o	–	–	–	o	o	o	o	–	o
	SF	++	o	++	o	–	–	–	o	o	o	++	o	+
	SP	++	o	o	o	–	–	–	o	o	o	+	–	+
	TD	++	o	o	o	o	–	–	o	o	o	o	–	+



**Fig. 15.1** Time series of emission density of sulphur ( $\text{mmol}_c \text{ S m}^{-2}$ ) in Germany, total deposition ( $\text{mmol}_c \text{ S m}^{-2}$ ) and  $\text{SO}_2$  concentration ( $\mu\text{g SO}_2 \text{ m}^{-3}$ ) in ambient air at Solling site (SO)

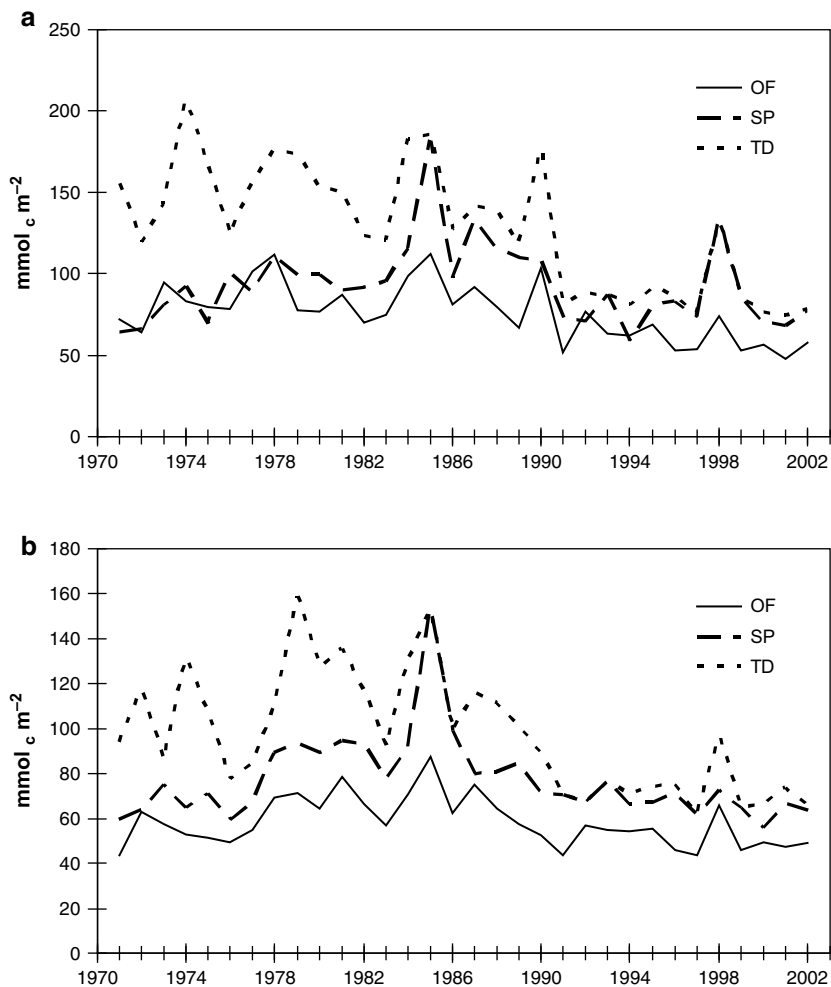
Mean  $\text{Na}^+$  fluxes in stand precipitation have been higher than in open field precipitation during the period 1990–2002 by a factor (ID/OF) of 0.49 at Göttinger Wald, 0.41 at Solling and 1.02 at Zierenberg sites. This factor ( $f_{\text{Na}}$ , see data collection and evaluation procedures) is used to calculate the particulate interception which is higher at the Zierenberg site than at the Solling and Göttinger Wald sites and may be caused by the high agricultural activity surrounding the Zierenberg site, and by higher aerodynamic roughness of the forest stand, which is situated at the slope of a relatively isolated mountain.

A high fraction of total acid inputs was contributed by  $\text{NH}_4^+$ , where the acid inputs are given by the sum of  $\text{H}^+$ ,  $\text{Mn}^{2+}$  and  $\text{NH}_4^+$  depositions. Despite decreasing  $\text{NH}_4^+$  fluxes at the Solling and Göttinger Wald sites during the last two decades, the relative contribution of  $\text{NH}_4^+$  to the fluxes of acids has increased significantly because of a considerable decrease in free acidity fluxes. During 1990–2002, the contribution of  $\text{NH}_4^+$  to total acid deposition was 50–85% at Solling, 75–95% at Göttinger Wald and 40–65% at Zierenberg, whereas at the beginning of the 1970s,  $\text{NH}_4^+$  contributed from 10 to 40% at the Solling site.

Annual element fluxes for open field deposition, throughfall and stemflow at Solling, Göttinger Wald, and Zierenberg are documented in Annex Tables 15.11–15.19.

## 15.4 Canopy Rain Interactions

Various tree compartments (leaves, twigs, branches and bark) act as sources or sinks for solutes in precipitation when they pass through the canopy. The canopy budget is commonly estimated by subtracting stand precipitation from total deposition

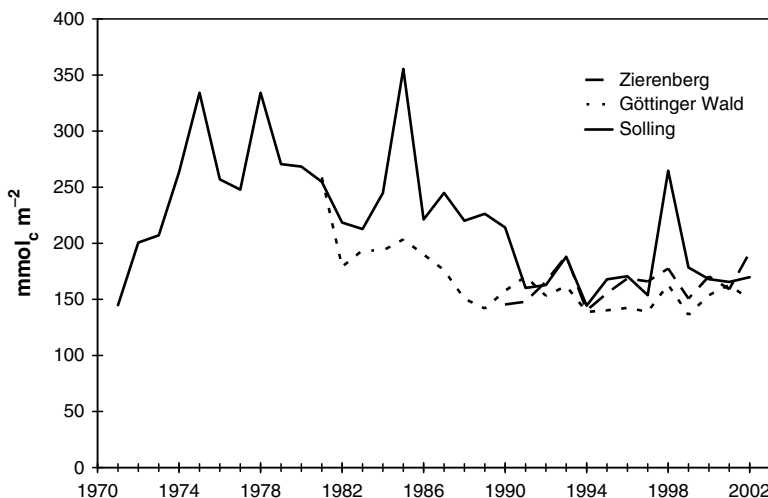


**Fig. 15.2** Time series of annual fluxes of (a)  $\text{NH}_4^+$  and (b)  $\text{NO}_3^-$  with open field deposition (*OF*), stand precipitation (*SP*) and total deposition (*TD*), according to Ulrich 1994) at Solling site

(canopy model by Ulrich 1994). Negative values can be interpreted as indication of leaching from the leaves (Langusch et al. 2003) and of dissolution of dry deposited particles, whereas positive values indicate a sink function, e.g. uptake by the canopy. Plant leaching may occur as a diffusion of organically complexed cations or as an exchange process, where nutrient cations such as  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$  and  $\text{Mn}^{2+}$  are exchanged against  $\text{H}^+$  or  $\text{NH}_4^+$  (Klemm et al. 1989; Draaijers and Erisman 2005).

Mean values for canopy budgets have been negative for  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$  and  $\text{Mn}^{2+}$ , whereas they have been positive for  $\text{H}^+$ ,  $\text{NH}_4^+$  and  $\text{NO}_3^-$  (Table 15.9). At the Zierenberg site, no retention of  $\text{NO}_3^-$  has been observed during the observation period. Retention of nitrogen by the canopy can be caused by (1) uptake by





**Fig. 15.3** Time series of annual  $N_{\text{tot}}$  fluxes with stand precipitation at the Solling, Göttinger Wald and Zierenberg sites

**Table 15.9** Average canopy budgets (total deposition—stand precipitation) for a number of elements during the periods 1969–1980 (Solling only, 1971–1980 for  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ), 1981–1989 (Solling and Göttinger Wald), and 1990–2002 at the three sites

Site	Period	$\text{K}^+$ $\text{Mg}^{2+}$ $\text{Ca}^{2+}$ $\text{H}^+$ $\text{Mn}^{2+}$ $\text{NH}_4^+$ $\text{NO}_3^-$						
		(mmol <sub>c</sub> m <sup>-2</sup> per year)						
Solling	1969–1980	-55	-5	-29	65	-11.8	70	36
	1981–1989	-56	-12	-39	103	-10.3	29	22
	1990–2002	-51	-6	-14	34	-3.7	12	6
Göttinger Wald	1981–1989	-50	-12	-46	88	-0.9	5	5
	1990–2002	-52	-6	-17	29	-0.2	6	7
Zierenberg	1990–2002	-65	-10	-5	50	-0.1	18	0

epiphytic algae and lichens, (2) immobilisation by micro-organisms (Stadler and Michalzik 2000), and (3) assimilation into the leaves (Garten et al. 1998). The last process is likely to be the most relevant one for the retention of N by the canopy (Lovett and Lindberg 1993).

Mean  $\text{NH}_4^+$  retention rates by the canopy decreased during the study period at the Solling site. For the measurement period of 1990–2002,  $\text{NH}_4^+$  retention by the canopy was estimated as: 12 mmol<sub>c</sub> for Solling, 6 mmol<sub>c</sub> for Göttinger Wald, and 18 mmol<sub>c</sub> m<sup>-2</sup> per year for Zierenberg. The corresponding figures for  $\text{NO}_3^-$  were: 6 mmol<sub>c</sub> (Solling), 7 mmol<sub>c</sub> (Göttinger Wald) and 0 mmol<sub>c</sub> m<sup>-2</sup> per year (Zierenberg), and for  $N_{\text{tot}}$ : 18 mmol<sub>c</sub> (Solling), 13 mmol<sub>c</sub> (Göttinger Wald) and 18 mmol<sub>c</sub> m<sup>-2</sup> per year (Zierenberg). As mentioned above, these estimates are uncertain and are most probably an underestimation. For the sites of the Integrated Forest Study (IFS), mean inorganic N-retention rates of 10–160 mmol<sub>c</sub> m<sup>-2</sup> per year were found (Lovett and Lindberg 1993), which brackets the values found for the sites of



**Fig. 15.4** Canopy retention (N-uptake) of total N compared to requirement for growth increment and forest requirement (growth and turnover) at Solling, Göttinger Wald and Zierenberg (data for N increment and N requirement from Rademacher et al. (Chap. 8))

this study. Horn et al. (1989) calculated N-retention rates by the canopies of a healthy and a declining spruce stand at Fichtelgebirge, Bavaria, of 89 and 185  $\text{mmol}_c \text{m}^{-2}$  per year, respectively.

If all the assumptions for calculating the canopy retention of N were valid, the canopy absorption would cover 7% of N-demand for growth and turnover at the Solling stand which has been calculated to be 740  $\text{mmol m}^{-2}$  (Chap. 8). Similar values of N-demand calculated for the Göttinger Wald (770  $\text{mmol m}^{-2}$  per year) and Zierenberg (360  $\text{mmol m}^{-2}$  per year) stands (Chap. 8), of which about 1.5% and 5% will be taken up by the canopy from atmospheric deposition (Fig. 15.4). Compared to the N required for the growth increment, the Solling stand can cover its N-demand completely by uptake within the canopy, whereas the relative contribution of crown uptake is low for the Göttinger Wald (11%) and Zierenberg stands (25%). Annual N-retention for forest growth has been estimated as 43  $\text{mmol m}^{-2}$  for the stands at Solling, 38–63  $\text{mmol m}^{-2}$  for Göttinger Wald and 126  $\text{mmol m}^{-2}$  for Zierenberg (Chap. 8).

The assimilation of  $\text{NH}_4^+$  and  $\text{NO}_3^-$  from precipitation has been experimentally confirmed by Brumme et al. (1992); Veithen (1996); Garten et al. (1998); and Harrison et al. (2000); Brumme et al. (1992) found that 6–12% of  $^{15}\text{N}$  applied to the aboveground parts of 3 to 9-years-old beech plants from the Solling site over 4 months was allocated to the roots. By the use of  $^{15}\text{N}$  labelled  $\text{NH}_4^+$  and  $\text{NO}_3^-$ , they found higher uptake rates for  $\text{NH}_4^+$  as compared to  $\text{NO}_3^-$  indicating preferential uptake of  $\text{NH}_4^+$ . In a similar study, Garten et al. (1998) found a retention of  $^{15}\text{N}$  labelled wet deposition of 12–26% for deciduous trees at Walker Branch Watershed, Tennessee, USA. Veithen (1996) used washing procedures at leaves from the Solling and Göttinger Wald stands for the study of canopy interactions and also found higher uptake rates for  $\text{NH}_4^+$  as compared to  $\text{NO}_3^-$ . The preferential uptake of  $\text{NH}_4^+$  is confirmed for the Solling and Zierenberg sites from the data of this study.

At the Solling site, uptake of  $\text{NH}_4^+$  and  $\text{NO}_3^-$  by the canopy decreased during the last 18 years (Table 15.10). At the Göttinger Wald and Zierenberg sites, no such

**Table 15.10** Trends of canopy budgets at Solling and Göttinger Wald sites from 1981 to 2002 and at Zierenberg site from 1990 to 2002 (test of trend with correlation coefficient after Pearson; o no trend, + significant increasing trend  $p \leq 0.05$ , ++ highly significant increasing trend  $p \leq 0.01$ , – significant decreasing trend  $p \leq 0.05$ , – – highly significant decreasing trend  $p \leq 0.01$ )

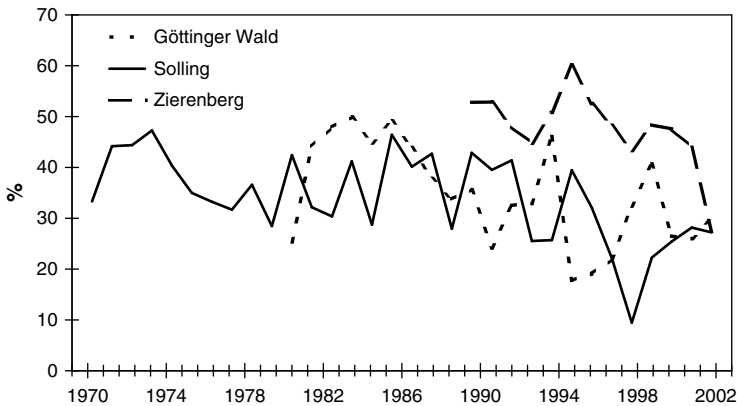
Site	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>
Solling	o	+	++	– –	++	–	–
Göttinger Wald	o	++	++	– –	++	o	o
Zierenberg	o	o	o	– –	o	o	o

trend was observed. Brumme et al. (1992) and Veithen (1996) showed that NH<sub>4</sub><sup>+</sup> uptake is dependent on the NH<sub>4</sub><sup>+</sup> concentration in precipitation. Thus, decreasing uptake rates of NH<sub>4</sub><sup>+</sup> may be attributed to decreasing NH<sub>4</sub><sup>+</sup> concentrations in deposition. Veithen (1996) estimated the compensation point for NH<sub>4</sub><sup>+</sup>, where NH<sub>4</sub><sup>+</sup> uptake turns to NH<sub>4</sub><sup>+</sup> leaching, to be between 28 and 46 μmol<sub>c</sub> l<sup>-1</sup>. As mean NH<sub>4</sub><sup>+</sup> concentrations in throughfall and stemflow at the Solling and Göttinger Wald sites are currently well above the compensation point found by Veithen (1996), NH<sub>4</sub><sup>+</sup> uptake is likely to take place at these stands.

Positive canopy budgets of H<sup>+</sup> indicated proton buffering at all sites. The buffering can occur as an exchange process where H<sup>+</sup> is exchanged against cations such as K<sup>+</sup>, Mg<sup>2+</sup> or Ca<sup>2+</sup> (Lindberg et al. 1986; Bredemeier 1988; Klemm et al. 1989; Matzner and Meiwes 1994; Draaijers and Erisman 1995). Mean proton buffering rates for 1990–2002 were calculated at 34 mmol<sub>c</sub> (Solling), 29 mmol<sub>c</sub> (Göttinger Wald) and 50 mmol<sub>c</sub> m<sup>-2</sup> per year (Zierenberg). Between 1981 and 2002, proton buffering rates decreased significantly at the Solling and Göttinger Wald sites accompanied by a simultaneous decrease in the leaching rates of Mg<sup>2+</sup> and Ca<sup>2+</sup> (Table 15.10). Proton buffering rates decreased significantly at the Zierenberg site between 1990 and 2002.

Buffering of total acidity in the canopy has been calculated from the sink functions of H<sup>+</sup> and NH<sub>4</sub><sup>+</sup>. At the study sites, 13–59% of the load of total acidity is buffered in the canopy on an annual basis (Fig. 15.5). In addition to decreasing loads of total acidity, the degree of buffering of acidity in the canopy has also decreased during the last 22 years. For the Göttinger Wald site, this might be explained by the more or less constant NH<sub>4</sub><sup>+</sup> uptake, which causes a production of H<sup>+</sup> in the canopy (Ulrich 1994). At the Solling site, the decrease of H<sup>+</sup> buffering has been twice as high as the decrease of NH<sub>4</sub><sup>+</sup> uptake. Hence, the H<sup>+</sup> production by NH<sub>4</sub><sup>+</sup> uptake has become more important. At the Zierenberg site, the degree of acid buffering in the canopy was generally higher compared to the other sites that also decreased during the study period (Fig. 15.5).

Leaching rates of K<sup>+</sup> from the canopy have been similar at the three study sites, whereas leaching of Mn<sup>2+</sup> has occurred only at the Solling site probably due to the higher Mn<sup>2+</sup> availability in this acid soil (Table 15.9). The higher amount of leaching of Mn<sup>2+</sup> from the canopy at the Solling site was confirmed by Veithen (1996). Leaching of Mg<sup>2+</sup> was highest at the Zierenberg site because of high Mg foliar content and also high Mg<sup>2+</sup> concentration in soil solution resulting from the high Mg<sup>2+</sup> content of the magmatic bedrock. Leaching of Ca<sup>2+</sup> from the canopy was



**Fig. 15.5** Time series of buffering of acids in the canopy relative to total acidity load at the Zierenberg, Solling and Göttinger Wald sites

similar at the Solling and Göttinger Wald sites, but lower at the Zierenberg sites. Contrary to these results, Veithen (1996) found higher leaching rates for  $\text{Ca}^{2+}$  from the canopy at the Göttinger Wald site than at the Solling site, which were attributed to a better  $\text{Ca}^{2+}$  nutrition of the stand growing on calcareous substrate. Surprisingly, the  $\text{Ca}^{2+}$  availability of the soils and also foliar contents of the three sites were not reflected by the  $\text{Ca}^{2+}$  leaching rates from the canopy. Mohr et al. (2005) found a positive relation between leaching rates of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  and foliar nutrient concentrations. Time series of canopy budgets indicate that leaching rates of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{Mn}^{2+}$  decreased significantly during 1981 and 2002 at the Solling and Göttinger Wald sites (Table 15.10). A decrease in the leaching rates of nutrient cations from the canopy may be related to decreasing deposition rates of  $\text{H}^+$ . No significant trends of canopy budgets have been observed at the Zierenberg site for these ions.

## 15.5 Discussion

Atmospheric deposition of many major elements can be attributed almost quantitatively to human activities. The only important natural source of salts is of marine origin. Main sources of anthropogenic air pollutants are: combustion processes, industrial processes and agriculture. Combustion and industrial processes are mainly responsible for the emission of  $\text{SO}_2$  and  $\text{NO}_x$ , whereas dust particles are released mainly through industrial processes.  $\text{NH}_3$  is mainly emitted through agricultural activities related to animal farming (Umweltbundesamt 2000). Deposition rates at the study sites are moderate when compared to other study sites in Germany (Gauger et al. 2002), but are relatively high compared to other regions in Europe (Hauhs et al. 1989; de Vries et al. 2001). Compared to spruce stands, deposition at beech stands is generally lower (Meessenburg et al. 1995; Balazs 1998; Rothe et al. 2002, Matzner et al. 2004); Eichhorn et al. (2001) evaluated data from 49 beech

plots across Europe with average total N-deposition of  $136 \text{ mmol}_c \text{ m}^{-2}$  per year (min–max:  $59\text{--}210 \text{ mmol}_c \text{ m}^{-2}$  per year). The study sites are with  $151\text{--}179 \text{ mmol}_c \text{ m}^{-2}$  per year among the European beech sites with above-average N-deposition. Compared to 144 intensive monitoring plots in Europe,  $\text{Na}^+$  scaling factors ( $f_{\text{Na}}$ ) at the study sites are above average (medium value 0.34, de Vries et al. 2001), suggesting that the contribution of dry deposition to total deposition is more important than at most other forests in Europe.

Deposition rates of the most major elements decreased during the last two decades which is evident at the Solling and Göttinger Wald sites with long observation periods. Atmospheric deposition is primarily characterised by  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ . During the 1970s and 1980s,  $\text{H}^+$  also played a major role in precipitation chemistry. During the last two decades deposition of free acidity has decreased but that of potential acidity has remained high due to high  $\text{NH}_4^+$  deposition. The reductions of  $\text{SO}_4^{2-}$  deposition are related to reductions of  $\text{SO}_2$  emission in western Germany (Umweltbundesamt 2000). Because  $\text{H}^+$  is mainly generated through the oxidation of  $\text{SO}_2$  to  $\text{SO}_4^{2-}$ , the  $\text{H}^+$  deposition decreased simultaneously. The reduction of acid depositions to central German forest ecosystems is attributed to the reduction of emissions, which became effective through legislation and the closing down of industrial units in eastern Germany after the reunion. There has been a reduction in dust emissions, decreasing the depositions of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ . This decrease in cations may have consequences for the buffering of acids in precipitation and for the nutrition of the forest stands. N-deposition decreased only slightly, which is also in agreement with constantly high N-emissions in western Germany (Umweltbundesamt 2000). N-deposition at the three sites is currently well above the amount retained for the forest increment.

Estimates of total deposition of different N-species are very uncertain because of the involvement of several different deposition and transformation processes in the canopy (Marques et al. 2001). N-deposition into forests occurs as rainwater, as fog or in gaseous form and with several different N-species. The deposited N-compounds interact with the canopy including their assimilation into leaves and transformations of the N-species. Garten et al. (1998) argued that the assimilation of gaseous N-compounds was the most effective uptake process, whereas Harrison et al. (2000) estimated the uptake from wet deposition to be more important. N-leaching from the canopy is possible at certain growth phases. Due to the complex biochemical processes, estimates of total N-deposition with a simple approach such as the model of Ulrich (1994) should be viewed with due care. Because N-uptake and gaseous deposition cannot be independently calculated, an underestimation of total deposition is likely by this model. Alternative canopy budget models are more specific with respect to nitrogen, but require independent input parameters, which are sometimes difficult to estimate (Horn et al. 1989; Draaijers and Erisman 1995). Independent measurements with micro-meteorological methods at a spruce stand adjacent to the Solling beech stand suggested an underestimation of N-deposition of almost 50% by the Ulrich model (Ibrom et al. 1995; Marques et al. 2001). Gauger et al. (2002) compared deposition estimates from inferential modelling with estimates from throughfall measurements and canopy

budget modelling and found an underestimation of about 50% by canopy budget modelling for Level II monitoring plots in Germany stocked with spruce. For Swiss long-term forest monitoring sites, Schmitt et al. (2005) found on an average  $17 \text{ mmol}_c \text{ m}^{-2}$  per year higher N-deposition rates estimated with an inferential method than by throughfall measurements. Nevertheless, estimates of total deposition for the study sites are plausible but are regarded as lower limits of true values.

Acid deposition on the canopy will induce interaction between rainwater and foliage. Leaching of nutrients from the canopy may induce nutrient deficiencies depending on the capacity of the trees to replenish the nutrient pools (Lindberg et al. 1986). When large quantities of N are assimilated in the canopy, the N-uptake by roots may be reduced. This may increase nutrient imbalances due to the spatial decoupling of N and base cation uptake (Harrison et al. 2000). Finally, the major changes in the deposition have also affected canopy-rain interactions. A decrease in the atmospheric emissions has reduced interaction between precipitation and canopy. As precipitation acidity is a major driving force for exchange of nutrient cations at foliage surfaces and their subsequent leaching, a reduced acidity input results in decreased exchange rates (Klemm 1989).

## 15.6 Conclusions

- N concentrations in open field precipitation, throughfall and stemflow at three beech stands in the northwest German low mountain ranges differ substantially between sites and pathways. However, the contributions of the different N-species  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and  $\text{N}_{\text{org}}$  to the deposition fluxes are relatively similar between sites.
- The sources of solutes in the deposition pathways can be related to anthropogenic air pollution, sea spray, deposition of organic compounds and canopy processes.
- Atmospheric deposition of most major components has declined over the last two decades. Reductions of deposition of acids and S have been more pronounced than reduction of N-input.
- Estimation of total N-deposition with the canopy budget model of Ulrich (1994) most probably underestimates real input rates. However, total deposition estimates can be regarded as lower limits of true values.
- According to the canopy budget model, N is taken up by the canopy of the beech stands. The uptake of  $\text{NH}_4^+$  is higher than the uptake of  $\text{NO}_3^-$ .
- The N-requirement of beech stands for growth increment may be fulfilled to a substantial part by canopy uptake.
- Due to the decline of atmospheric deposition, the interactions between rainwater and the canopy have also reduced.

## Annex Tables

See Tables 15.11 to 15.19.

Table 15.11 Rates of open field deposition at Solling during the period 1969–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Year	H <sub>2</sub> O (mm)	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	(mmol <sub>c</sub> m <sup>-2</sup> )						N <sub>org</sub>	
							Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>	N <sub>tot</sub> (mmol m <sup>-2</sup> )		
1969	1,064	28	7.2	15.6	41	68.5	0.7	150	44					
1970	1,479	37	7.4	10.7	68	109.1	1.1	170	58					
1971	810	26	11.8	32.1	71	70.4	0.7	138	36	72	44	135	19	
1972	910	22	9.2	14.8	109	91.3	0.4	157	30	64	63	183	54	
1973	1,037	53	10.0	16.5	38	99.2	2.6	132	72	95	57	165	13	
1974	1,235	32	6.1	11.5	32	65.5	1.8	145	49	83	53	164	29	
1975	884	28	8.2	10.7	37	82.4	1.1	158	40	80	51	166	35	
1976	688	43	9.0	14.8	40	60.5	1.1	143	55	79	49	163	35	
1977	897	42	14.3	18.1	58	84.3	3.3	158	56	101	55	198	42	
1978	950	39	11.5	14.0	47	75.4	1.1	150	61	111	69	223	43	
1979	845	21	6.7	10.7	34	85.3	1.1	122	30	78	71	188	39	
1980	1,018	30	8.4	13.2	35	68.5	0.7	152	39	77	64	169	28	
1981	1,544	43	13.3	18.1	46	89.3	2.9	140	50	87	79	208	42	
1982	867	29	7.5	12.0	52	64.5	2.5	146	36	70	66	154	17	
1983	1,029	44	10.6	12.2	37	61.5	1.3	131	54	75	57	156	24	
1984	1,217	35	10.3	10.6	38	126.0	1.0	135	76	99	71	202	33	
1985	1,071	30	10.0	9.5	47	85.3	1.6	145	38	112	87	209	17	
1986	1,209	36	7.8	8.5	35	114.1	0.6	99	48	81	62	195	52	
1987	1,204	32	6.8	7.0	25	58.5	0.6	122	39	92	75	207	40	
1988	1,214	46	8.0	11.4	24	57.6	1.8	101	46	80	64	157	15	
1989	949	32	5.4	9.0	23	41.7	0.8	85	40	67	57	138	15	
1990	1,040	51	11.5	13.0	22	14.9	0.8	78	42	103	52	181	26	
1991	837	40	5.0	9.7	22	7.9	1.0	56	30	52	43	103	9	
1992	1,219	62	8.3	12.2	29	5.0	0.7	83	41	77	57	154	20	
1993	1,198	56	5.9	11.2	24	6.3	0.4	72	33	63	55	136	15	

(continued)

Table 15.11 (continued)

Year	H <sub>2</sub> O (mm)	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	(mmol <sub>c</sub> m <sup>-2</sup> )					N <sub>tot</sub> (mmol m <sup>-2</sup> )	N <sub>org</sub>
							Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>		
1994	1,386	41	4.9	8.4	16	20.5	0.5	69	38	62	54	125	8
1995	1,321	46	4.6	7.4	12	25.8	0.5	69	46	69	55	131	8
1996	928	26	2.8	4.2	10	17.9	0.3	49	23	53	46	103	2
1997	1,008	29	3.1	5.2	13	17.7	0.4	48	28	54	43	105	7
1998	1,575	39	5.9	6.3	15	27.8	0.6	69	39	74	66	147	7
1999	1,109	37	4.6	5.3	11	18.0	0.4	44	33	53	46	106	7
2000	1,131	33	4.6	4.4	10	16.6	0.4	46	28	57	49	119	13
2001	1,187	29	3.3	4.5	10	19.5	0.9	41	27	48	48	112	17
2002	1,451	27	4.5	4.3	10	25.3	0.2	46	28	58	49	122	15



Table 15.12 Throughfall fluxes at Solling during the period 1969–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Year	H <sub>2</sub> O (mm)	(mmol <sub>c</sub> m <sup>-2</sup> )										N <sub>org</sub>				
		Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>		N <sub>tot</sub> (mmol m <sup>-2</sup> )			
1969	793	40	46.8	24.7	104	77.4	10.9	195	61							
1970	1,035	51	46.8	28.0	129	113.1	8.4	258	86							
1971	541	42	32.7	30.5	106	90.3	8.4	179	61	58	52	129	19			
1972	623	31	47.1	22.2	126	76.4	6.6	202	51	52	54	159	54			
1973	759	67	46.8	28.8	93	83.3	10.9	188	95	77	69	185	39			
1974	832	48	66.5	26.3	85	60.5	16.4	144	74	76	57	191	58			
1975	632	44	47.8	23.0	87	89.3	10.9	204	63	48	60	203	81			
1976	550	50	76.0	29.6	96	79.4	10.6	247	78	91	51	229	86			
1977	745	47	47.1	25.5	93	116.1	15.3	231	68	76	56	204	72			
1978	770	44	49.4	26.3	93	100.2	12.7	238	67	100	79	271	92			
1979	710	35	28.1	18.9	65	84.3	7.3	180	35	85	80	221	56			
1980	762	43	45.5	23.9	74	85.3	10.6	220	65	84	76	216	56			
1981	1,050	53	46.6	26.3	82	83.3	13.1	202	64	79	81	201	41			
1982	562	37	39.0	22.8	97	51.4	8.5	187	59	81	84	185	20			
1983	652	54	52.1	22.6	70	46.4	9.6	151	80	78	64	164	21			
1984	832	50	57.7	25.3	81	93.7	11.8	222	69	102	78	210	30			
1985	692	39	56.6	27.2	100	83.3	13.3	238	70	156	133	303	24			
1986	838	42	39.0	18.6	68	56.9	6.5	150	60	79	73	170	19			
1987	795	38	72.7	24.2	76	35.4	11.3	208	58	120	67	212	26			
1988	844	57	42.7	23.7	71	37.3	8.9	165	78	104	69	186	26			
1989	647	41	45.9	19.2	52	24.6	7.0	122	55	95	73	191	33			
1990	737	62	74.7	23.9	49	17.8	6.2	119	67	95	60	180	25			
1991	603	54	46.2	18.8	50	10.3	7.4	97	56	64	61	140	16			
1992	834	66	77.7	21.3	47	8.8	4.5	114	58	68	64	154	22			
1993	842	57	31.5	17.5	42	10.8	3.7	100	44	82	69	171	21			

(continued)

Table 15.12 (continued)

Year	H <sub>2</sub> O (mm)	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>	N <sub>tot</sub> (mmol m <sup>-2</sup> )	N <sub>org</sub>
(mmol <sub>c</sub> m <sup>-2</sup> )													
1994	889	44	28.8	14.1	29	26.0	2.9	83	46	53	58	127	16
1995	863	51	85.8	16.1	25	17.1	4.2	106	59	73	58	148	16
1996	652	34	36.2	10.8	24	14.5	3.8	84	38	73	61	146	12
1997	690	35	30.7	10.3	24	12.3	3.3	63	40	66	54	136	15
1998	1,068	47	48.7	13.4	26	12.6	3.6	94	53	126	64	246	56
1999	736	41	39.4	10.1	20	10.3	3.7	50	44	75	54	153	25
2000	747	35	57.7	10.1	17	10.7	2.3	54	39	63	48	149	37
2001	912	37	30.8	9.6	22	11.5	3.6	54	42	62	58	146	26
2002	982	30	44.2	7.8	18	8.9	2.6	53	36	69	56	150	25

Table 15.13 Stemflow fluxes at Solling during the period 1969–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Year	H <sub>2</sub> O (mm)	(mmol <sub>c</sub> m <sup>-2</sup> )										N <sub>org</sub>			
		Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>		N <sub>tot</sub> (mmol m <sup>-2</sup> )		
1969	138	16	27.4	7.4	30	77.4	3.6	180	18						
1970	92	12	22.3	7.4	25	52.6	2.5	88	21						
1971	105	13	13.6	4.1	23	49.6	1.8	74	15	6	7	16	3		
1972	118	10	17.4	4.9	34	32.7	2.2	87	29	14	10	41	17		
1973	134	12	25.8	8.2	12	37.7	2.9	79	20	4	6	22	11		
1974	160	31	35.8	14.0	43	66.5	7.3	268	38	17	8	73	48		
1975	115	15	28.9	8.2	35	47.6	5.8	130	23	21	11	131	41		
1976	89	17	18.7	8.2	24	30.8	2.2	56	37	10	8	29	21		
1977	116	18	24.3	7.4	27	54.6	3.3	112	24	13	11	44	20		
1978	112	17	18.2	5.8	22	51.6	2.5	90	22	11	10	63	41		
1979	112	13	21.2	7.4	26	49.6	2.9	97	33	14	14	50	22		
1980	162	16	27.1	8.2	32	55.6	3.6	120	23	15	14	53	24		
1981	199	21	39.1	9.9	35	64.5	5.1	145	27	11	13	54	29		
1982	66	14	13.4	5.2	18	27.0	2.0	58	20	10	9	34	14		
1983	136	18	23.7	8.3	34	45.6	3.2	93	25	17	14	49	19		
1984	127	15	16.8	6.7	21	57.5	2.3	68	21	14	14	35	7		
1985	101	10	12.1	5.2	19	42.7	2.4	77	16	26	20	52	6		
1986	125	14	16.2	6.1	21	53.6	2.0	81	22	18	26	51	7		
1987	119	12	19.7	6.4	21	34.7	2.4	74	17	13	12	33	17		
1988	126	23	23.3	8.1	24	38.7	3.2	81	29	12	12	34	22		
1989	97	15	18.0	6.8	20	31.7	2.5	60	22	14	12	35	20		
1990	110	25	19.5	7.4	20	30.8	2.8	61	31	13	11	34	20		
1991	91	9	8.1	3.2	8	6.0	0.6	25	10	9	9	20	3		
1992	72	5	4.4	1.4	3	4.0	0.2	12	6	3	4	9	3		
1993	140	9	7.6	2.3	6	8.4	0.5	22	7	6	8	17	4		

(continued)

Table 15.13 (continued)

Year	H <sub>2</sub> O (mm)	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>	N <sub>tot</sub> (mmol m <sup>-2</sup> )	N <sub>org</sub>
1994	133	10	6.0	2.2	4	10.0	0.3	19	10	6	8	17	4
1995	129	11	14.7	2.5	4	7.1	0.4	23	13	8	8	20	4
1996	98	8	14.0	2.2	5	9.9	0.5	28	10	10	10	24	5
1997	104	6	7.0	1.4	4	5.0	0.3	14	6	7	8	18	3
1998	160	10	10.7	1.8	4	5.0	0.3	16	11	7	8	19	4
1999	110	9	8.6	1.7	3	4.6	0.3	14	9	10	11	25	4
2000	112	9	8.8	1.6	3	3.6	0.2	11	11	6	8	19	5
2001	137	8	11.1	1.4	3	2.1	0.3	11	9	6	8	20	5
2002	147	7	10.6	0.9	2	2.0	0.2	10	9	8	7	20	5

Table 15.14 Rates of open field deposition at Göttinger Wald during the period 1981–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Year	H <sub>2</sub> O (mm)	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	(mmol <sub>c</sub> m <sup>-2</sup> )						N <sub>tot</sub> (mmol m <sup>-2</sup> )	N <sub>org</sub>
							SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Mn <sup>2+</sup>		
1981	1,252	35	20.2	17.6	97	52.5	0.7	160	50	73	62	169	34	
1982	550	18	8.6	9.6	46	20.4	0.7	82	23	43	41	112	28	
1983	635	32	7.1	10.4	43	31.9	0.6	96	32	59	51	129	23	
1984	754	21	6.2	11.3	55	49.4	0.8	113	25	59	59	148	30	
1985	621	22	8.9	9.4	49	26.2	0.5	92	27	68	51	142	23	
1986	784	25	6.9	9.9	45	43.7	0.4	100	32	58	66	151	28	
1987	900	26	9.3	13.0	76	54.8	0.3	129	33	75	73	162	15	
1988	763	45	7.4	15.9	58	45.3	0.4	118	45	66	84	155	15	
1989	495	24	3.4	9.5	36	38.2	0.2	85	27	51	58	99	5	
1990	546	39	3.0	11.3	32	19.6	0.3	70	30	53	54	115	9	
1991	496	37	3.2	11.2	35	12.4	0.2	67	29	51	54	113	8	
1992	686	38	3.6	12.5	37	17.0	0.3	84	31	67	66	142	9	
1993	715	30	3.6	8.2	24	3.4	0.3	53	15	46	43	102	12	
1994	758	18	2.8	4.4	15	17.5	0.3	52	15	49	44	96	13	
1995	598	20	4.1	4.8	11	0.8	0.2	42	21	42	35	82	5	
1996	589	14	2.7	3.2	10	1.2	0.2	36	12	42	37	85	5	
1997	664	21	2.8	8.2	25	9.9	0.4	44	18	44	40	89	5	
1998	893	21	2.7	4.7	14	21.6	0.3	47	19	52	50	109	7	
1999	635	18	2.6	3.7	11	11.7	0.2	31	14	42	38	87	6	
2000	719	18	3.3	4.1	14	8.1	0.3	32	13	39	37	85	9	
2001	718	18	2.7	4.0	11	11.7	0.1	29	17	35	36	83	12	
2002	868	18	3.2	4.4	15	14.7	0.1	39	21	51	45	107	9	

**Table 15.15** Throughfall fluxes at Göttinger Wald during the period 1981–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Year	H <sub>2</sub> O (mm)	Na	K	Mg	Ca	H	Mn	SO <sub>4</sub> <sup>2-</sup>	Cl	NH <sub>4</sub>	NO <sub>3</sub>	N <sub>tot</sub> (mmol m <sup>-2</sup> )	N <sub>org</sub>
(mmol <sub>c</sub> m <sup>-2</sup> )													
1981	843	34	40.0	26.5	124	27.7	1.7	185	52	92	69	188	32
1982	366	19	31.4	20.3	89	8.1	1.0	112	33	54	53	129	22
1983	428	30	44.1	22.7	78	14.0	1.5	119	48	61	53	132	18
1984	543	23	39.3	22.0	100	16.7	1.4	147	36	62	59	149	28
1985	417	22	36.2	21.1	89	14.6	1.5	127	39	63	60	144	21
1986	556	23	34.2	18.8	87	19.2	1.1	122	38	49	65	144	29
1987	621	22	53.0	20.3	89	20.1	1.0	146	39	66	53	127	13
1988	522	28	25.9	15.2	66	15.0	0.8	99	36	52	51	115	15
1989	351	23	34.7	18.6	71	10.4	0.9	95	33	64	47	109	9
1990	415	36	50.6	18.7	51	3.6	0.7	76	41	60	42	113	12
1991	329	32	31.0	17.2	57	2.6	0.8	73	34	65	50	129	15
1992	501	33	50.3	19.9	53	4.6	0.7	79	37	63	47	132	22
1993	590	34	26.2	15.3	47	5.2	0.5	77	28	59	55	134	20
1994	562	26	32.7	10.9	33	11.5	0.5	60	30	47	51	112	14
1995	502	29	65.7	18.4	35	6.9	0.6	65	35	58	45	117	14
1996	450	19	31.4	9.3	29	5.9	0.3	58	23	60	51	123	11
1997	463	26	32.8	10.6	33	4.4	0.3	48	31	54	50	116	12
1998	647	29	46.4	11.5	32	5.6	0.4	57	34	68	55	142	20
1999	497	28	36.3	9.5	27	6.0	0.3	42	33	51	52	116	14
2000	547	25	51.8	9.9	26	3.9	0.3	41	29	58	48	134	28
2001	612	27	47.1	10.9	31	4.1	0.3	45	35	68	52	145	25
2002	627	23	43.2	8.6	27	3.6	0.3	45	31	60	51	131	20

**Table 15.16** Stemflow fluxes at Göttinger Wald during the period 1981–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Year	H <sub>2</sub> O (mm)	(mmol <sub>c</sub> m <sup>-2</sup> )										N <sub>org</sub>	
		Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>		N <sub>tot</sub> (mmol m <sup>-2</sup> )
1981	183	14	24.1	8.1	41	53.6	0.5	120	25	33	27	70	10
1982	90	8	18.8	5.8	28	16.9	0.7	65	16	20	23	50	7
1983	132	22	37.6	12.3	49	43.7	0.9	124	39	22	30	61	10
1984	137	11	30.9	8.7	40	33.7	0.4	102	20	16	19	45	10
1985	91	9	24.3	6.7	28	25.8	0.4	78	16	27	24	59	9
1986	102	9	21.7	7.7	34	24.8	0.4	75	17	16	23	46	7
1987	159	10	31.6	7.2	35	32.7	0.3	85	17	20	23	49	6
1988	95	8	16.0	4.4	18	7.9	0.2	48	12	19	13	36	4
1989	86	9	13.9	4.2	18	12.9	0.2	43	13	15	16	33	5
1990	104	15	22.5	6.0	19	9.9	0.3	43	19	18	21	45	6
1991	71	8	15.4	3.4	12	5.0	0.1	31	11	20	15	41	6
1992	95	9	15.3	3.3	11	6.0	0.1	27	11	7	9	21	5
1993	103	6	13.2	2.1	8	5.4	0.1	26	6	12	12	28	5
1994	156	9	17.8	2.5	10	8.0	0.1	30	11	11	14	27	5
1995	86	8	15.8	2.0	6	3.2	0.1	21	10	10	9	23	3
1996	78	4	12.3	1.3	5	2.3	0.1	17	5	9	8	20	3
1997	90	5	10.9	1.1	4	2.0	0.0	14	6	10	10	23	3
1998	108	5	11.5	0.9	4	1.1	0.1	11	5	9	9	21	3
1999	80	5	8.7	1.0	4	1.3	0.1	9	6	8	9	20	3
2000	99	7	12.2	1.4	5	1.0	0.0	10	8	6	9	20	4
2001	91	4	14.3	1.5	4	0.2	0.0	11	5	7	6	18	5
2002	95	5	15.2	1.3	4	0.2	0.0	12	6	7	8	19	4

**Table 15.17** Rates of open field deposition at Zierenberg during the period 1990–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Year	H <sub>2</sub> O (mm)	(mmol <sub>c</sub> m <sup>-2</sup> )										N <sub>org</sub>	
		Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>		N <sub>tot</sub> (mmol m <sup>-2</sup> )
1990	630	29	10.3	18.7	39	19.9	0.2	51	17	32	34	78	11
1991	529	21	6.9	17.4	39	29.2	0.2	49	17	28	35	72	9
1992	742	34	9.1	17.6	37	34.2	0.3	63	25	44	37	91	10
1993	742	28	7.7	30.4	61	18.3	0.3	60	21	52	42	103	9
1994	780	35	14.5	34.1	67	16.7	0.3	57	29	61	39	111	10
1995	721	29	6.9	19.1	33	17.9	0.3	46	22	42	30	82	10
1996	688	17	5.7	15.4	28	14.4	0.2	43	17	45	37	91	9
1997	707	18	4.3	16.8	32	18.8	0.1	42	22	31	32	69	7
1998	890	28	6.3	18.8	35	18.3	0.2	42	27	35	36	80	9
1999	802	27	5.1	16.1	30	13.2	0.1	32	23	30	31	73	11
2000	800	25	7.5	18.4	35	10.5	0.1	39	23	45	42	98	11
2001	758	24	3.6	13.8	27	8.7	0.1	26	22	24	33	65	7
2002	981	23	5.0	14.8	27	8.9	0.2	36	21	41	40	89	9



**Table 15.18** Throughfall fluxes at Zierenberg during the period 1990–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Year	H <sub>2</sub> O (mm)	(mmol <sub>c</sub> m <sup>-2</sup> )										N <sub>org</sub>	
		Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>		N <sub>tot</sub> (mmol m <sup>-2</sup> )
1990	424	42	61.6	39.9	73	9.9	0.6	77	35	40	70	123	13
1991	355	35	55.5	44.6	79	6.9	0.7	83	36	42	64	122	16
1992	508	54	114.4	55.7	81	18.8	0.7	94	61	50	67	144	26
1993	497	46	51.3	49.7	89	5.5	0.4	96	51	68	80	168	19
1994	537	50	49.2	53.6	97	10.2	0.3	76	54	49	62	125	14
1995	483	57	70.0	58.2	79	6.5	0.7	88	64	50	61	141	30
1996	491	33	61.0	38.8	62	5.8	0.3	90	44	60	75	152	18
1997	495	42	48.2	42.1	68	11.5	0.5	76	62	55	80	152	18
1998	646	54	75.2	48.8	78	4.8	0.5	70	68	65	73	164	26
1999	555	48	46.0	33.8	57	7.024	0.3	49	57	48	71	135	16
2000	555	49	80.6	42.3	66	6.65	0.2	57	59	61	62	147	24
2001	549	46	66.7	34.4	54	2.383	0.4	51	59	43	65	128	20
2002	755	43	86.4	35.9	56	3.617	0.4	62	52	70	67	165	28

**Table 15.19** Stemflow fluxes at Zierenberg during the period 1990–2002 (Data source: Nordwestdeutsche Forstliche Versuchsanstalt)

Year	H <sub>2</sub> O (mm)	(mmol <sub>c</sub> m <sup>-2</sup> )										N <sub>org</sub>	
		Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> <sup>-</sup>		N <sub>tot</sub> (mmol m <sup>-2</sup> )
1990	58	7	10	5	10	3	0.1	17	5	9	11	22	3
1991	50	7	11	5	12	5	0.1	22	7	10	12	26	4
1992	68	8	12	5	10	14	0.1	18	8	8	10	22	4
1993	74	6	10	5	10	4	0.1	16	6	11	7	21	3
1994	79	6	8	5	10	3	0.0	11	7	8	4	15	3
1995	68	8	9	7	11	3	0.1	23	10	6	5	15	4
1996	70	5	10	4	8	2	0.1	19	6	7	6	17	4
1997	72	5	10	4	8	3	0.0	16	7	5	4	14	4
1998	96	8	14	5	9	1	0.1	15	10	5	3	14	6
1999	85	9	13	5	8	1	0.0	12	11	5	5	16	6
2000	83	11	19	5	10	1	0.0	14	14	10	7	23	7
2001	82	9	23	5	9	0	0.1	18	13	13	9	31	9
2002	113	7	19	4	6	0	0.0	12	9	14	6	27	8

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