4 SULPHATE AND NITROGEN DEPOSITION TO FORESTS AND TREND ANALYSES

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4.1 Introduction

The atmospheric deposition of sulphur (S) and nitrogen (N) compounds affects forest ecosystems through several processes. Deposition of acidifying compounds, inorganic nitrogen as a nutrient and base cations to forests in Europe is a major driver for many processes in forests.

In the frame of the LRTAP convention deposition measurements are carried out within both, the EMEP as well as in the ICP Forests programme of the WGE. One of the tasks of EMEP is a set of tools to derive maps of pollution levels and loads from emission inventories using transmission modelling. The bulk or wet deposition measurements of EMEP are used as validation for the maps in order to improve the tools.

The main objective of deposition measurements within ICP Forests are quantitative on-site (Level II) measurements of atmospheric deposition to forests across Europe usable for process oriented investigation of the causal chain between deposition and effects in forest ecosystems that are carried out on the ICP Forests Level II plots as well as assessing its changes over time. The objective of this chapter is to describe the bulk and throughfall deposition of sulphate and inorganic nitrogen (nitrate and ammonium) and its trends on ICP Forests Level II plots.

4.2 Methods

Continuous sampling of throughfall (TF) and bulk deposition (BD) is carried out on ICP Forests intensive monitoring plots (Level II) and at nearby open fields, respectively. The methods used in the countries (France: Ulrich & Lanier, 1993; Norway: Kvaalen et al., 2002; Moffat et al., 2002; Italy: Mosello et al., 2002; Switzerland: Thimonier et al., 2005; Finnland: Lindroos et al., 2006; Denmark: Gundersen et al., 2009; Czech Republic: Boháčová et al., 2010; Latvia: Lazdiņš, 2010; UK: Vanguelova et al., 2010; Swedish Throughfall Monitoring Network (SWETHRO): Pihl Karlsson et al., 2011; Belgium: Verstraeten et al., 2012) follow the ICP Forests Manual (earlier versions and ICP Forests, 2010). Collectors (approximately 10 to 20 replicates) are placed in the forest based on a random or fixed design in order to cover the spatial variation. Tests to determine the minimal number of samplers required to cover spatial variations to gain a representative plot mean have been carried out on a number of plots (e.g. Switzerland: Thimonier, 1998; UK: Houston et al., 2002; Belgum: Staelens et al., 2006). Samples are collected at least monthly,

\[ \text{For contact information, please refer to Annex III-4}. \]
filtered, and then stored at about 4°C before chemical analyses are performed to determine the concentrations of the macronutrients. The laboratory results are checked for internal consistency based on the conductivity, the ion balance, the concentration of organic N and the Na/Cl ratio, and analyses are repeated if suspicious concentrations are identified. The QA/QC procedures further include the use of control charts for internal reference material to check long-term comparability within national laboratories as well as participation in periodic working ring tests (e.g. Marchetto et al., 2006) to check international comparability.

Data was submitted annually by countries to the Programme Coordinating Centre (PCC), checked for consistency and stored in ICP Forests database.

Data from each sampling period were interpolated to regular monthly and annual data by: (i) splitting each sampling period overlapping two consecutive months and distributing precipitation quantity in proportion to the duration of the new sampling periods; (ii) setting deposition = 0 for periods with missing concentrations and mean precipitation < 0.1 mm day\(^{-1}\); (iii) calculating bulk and throughfall deposition (\(q_c\), in kg ha\(^{-1}\)) by multiplication of the precipitation quantity (\(q\), in L m\(^{-2}\)) and the concentrations (\(c\), in mg L\(^{-1}\)); (iv) summing up fluxes by months and years, respectively.

The current deposition has been determined for plots with continuous measurements during 2011. We analysed temporal trends for plots with continuous measurements from 2006 to 2011 (6 years). We used the following completeness criteria of continuous measurement: (i) sampling during more than 330 days per year, (ii) missing concentration values for less than 35 days per year. Sampling periods with mean precipitation below 0.1 mm day\(^{-1}\) were counted as non-missing even if no chemical analyses could be performed. Trend analyses were carried out with the Partial Mann-Kendall (PMK) tests (Libiseller & Grimvall, 2002) using interpolated monthly deposition data. Trend slopes were estimated following Sen (1968). These trend analyses were carried out in R (R Development Core Team, 2009) using the ‘rkt’ package (Marchetto, 2013).

### 4.3 Results

#### 4.3.1 Current deposition

Annual throughfall (TF) and bulk deposition (BD) of sulphur and nitrogen was calculated for 221 plots in 24 countries with continuous measurement in 2011 (Figure 4.3.1-1 and Figure 4.3.1-2). High sulphur deposition (Figure 4.3.1-1) has been measured in northern central Europe especially in a region covering Belgium/Netherlands, Central Germany, Czech Republic and Poland, reaching up to the southern Baltic and the Central Hungarian area. Furthermore, high values have also been observed in some Mediterranean regions in Spain, France, Southern Italy and Greece. High sulphur deposotions along the coast mostly occur with high Cl deposition, which is typical for sulphur that originates from sea salt.
Figure 4.3.1-1 Annual sulphate sulphur (SO$_4^-$-S) bulk and throughfall deposition in 2011 (in kg ha$^{-1}$).
Figure 4.3.1-2 Annual nitrate (NO$_3$-N) and ammonium (NH$_4$-N) bulk and throughfall deposition in 2011 (in kg ha$^{-1}$)
High nitrogen deposition (Figure 4.3.1-2) is also recorded in northern central Europe, as for sulphur, but extends further to the South down to southern Germany and the Swiss Plain and also further to the West, to northern France and central UK. In contrast to sulphur, the regions south of the Alps show relatively high bulk and throughfall deposition of nitrate and ammonium as well. In the Mediterranean area, relatively high values have been recorded at some sites in Spain.

4.3.2 Temporal trends

Trends for the period were calculated for 127 and 115 plots with continuous throughfall and bulk deposition measurements from 2006 to 2011, as well as for 83 and 78 plots with continuous throughfall and bulk deposition measurements from 2002 to 2011 (10 years), respectively (Figure 4.3.2-1, Figure 4.3.2-2 and Figure 4.3.2-3).

![Graph showing temporal trends of sulphate and nitrogen deposition](image-url)

**Figure 4.3.2-1:** Mean sulphate ($\text{SO}_4^{2-}$-S) and inorganic nitrogen ($\text{NO}_3^-$-N + $\text{NH}_4^+$-N) bulk and throughfall deposition on plots with continuous deposition measurements from 2002 to 2011 (n=number of plots).

The mean of the annual deposition of plots with continuous measurements from 2002 to 2011 decreased from about 8 and 6 kg S ha$^{-1}$ a$^{-1}$ to about 6 and 5 kg S ha$^{-1}$ a$^{-1}$ for throughfall and bulk...
deposition, respectively (Figure 4.3.2-1). The sulphur deposition showed a decreasing trend on the majority of plots for both periods 2002 to 2011 as well as 2006 to 2011 (Figure 4.3.2-2).

**Figure 4.3.2-2** Trend of sulphate ($\text{SO}_4^{2-}$) throughfall deposition on plots with continuous measurements from 2002 to 2011 and from 2006 to 2011. Non-significant positive trends are indicated with ‘no change (+)’ and non-significant negative trends with ‘no change (-)’

The mean of annual throughfall deposition of inorganic nitrogen on plots with continuous measurements from 2002 to 2011 decreased from about 13 to about 11 kg N ha$^{-1}$ a$^{-1}$ in 2011 (Fig. 4.3.2-1); however, this decrease is not statistically significant. For nitrogen significant decreasing trends have been detected for fewer plots than for sulphate sulphur (Fig. 4.3.2-3).
Figure 4.3.2-3: Trend of nitrate (NO$_3$-N) and ammonium (NH$_4$+-N) throughfall deposition of plots with continuous measurements from 2002 to 2011 and 2006 to 2011. Non-significant positive trends are indicated with ‘no change (+)’ and non-significant negative trends with ‘no change (-)’.
4.4 Discussion

The patterns of atmospheric N and S deposition of the year 2011 as well as the trends of bulk and throughfall deposition for the periods 2002 to 2011 and 2006 to 2011 show similar patterns than that of ICP Forests deposition measurements in earlier years. The trends are also in agreement with other studies of either national ICP Forests data, EMEP data or other deposition measurements (e.g. Meesenburg et al. (1995), Rogora et al. (2006), Staelens et al. (2012), Pihl Karlsson et al. (2011), Kvaalen et al. (2002), Vanguelova et al. (2010), Graf Pannatier et al. (2011), Marchetto et al. (2013), Verstraeten et al. (2012), Johnson et al. (2013), Hunova et al. (2004), Oulehle et al. (2011), Fagerli et al. (2008)).

The atmospheric deposition values presented here are restricted to bulk and throughfall deposition fluxes of inorganic compounds. The total deposition to forests also includes organic compounds, stemflow, as well as canopy uptake. Especially for nitrogen, the exchange in the canopy causes the total deposition to typically be different from the throughfall fluxes.

4.5 Conclusions

Atmospheric deposition of N and S compounds to forests at the ICP Forests Level II plots cover a relatively wide range and are still relatively high at certain plots. There is a main tendency of decreasing atmospheric depositions in the last 6 and 10 years especially for S compounds. However, trend slopes vary from plot to plot. Significant decreasing trends have not been observed for all plots and especially for nitrogen compounds there were plots with significant increasing trends as well, especially for the period 2006 to 2011.

4.6 References


Forest Condition in Europe
2013 Technical Report of ICP Forests

Report under the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP)

Alexa Michel, Walter Seidling, Martin Lorenz, Georg Becher (Eds.)

Thünen Working Paper 19