

**CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION  
INTERNATIONAL CO-OPERATIVE PROGRAMME ON ASSESSMENT AND MONITORING  
OF AIR POLLUTION EFFECTS ON FORESTS  
and  
EUROPEAN UNION SCHEME  
ON THE PROTECTION OF FORESTS AGAINST ATMOSPHERIC POLLUTION**

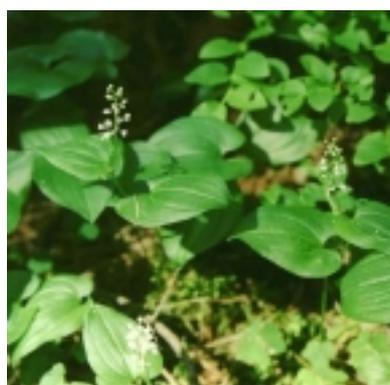
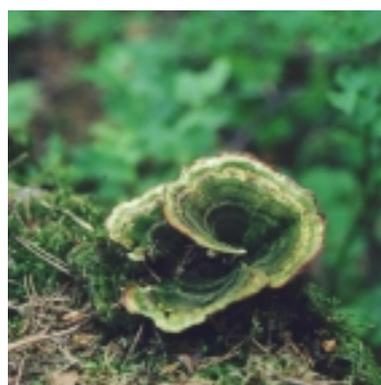
United Nations  
Economic Commission  
for Europe

European Commission

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# **Intensive Monitoring of Forest Ecosystems in Europe**

Technical Report 1999



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# **Intensive Monitoring of Forest Ecosystems in Europe**

Technical Report 1999

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**Forest Intensive Monitoring Coordinating Institute, 1999**



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## Abstract

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In this Technical Report, an overview is given of (i) the implementation of the 'Pan-European programme for Intensive Monitoring of forest ecosystems' until present, (ii) methodological aspects for the evaluation, (iii) results of key parameters in deposition, meteorological and soil solution surveys and (iv) relationships between key parameters in the deposition and soil solution surveys and various site and stand characteristics, the latter being derived from data in the forest growth (increment) survey. The following major interesting results were found:

- On average, the total nitrogen input is 50% larger than the total sulphur input. N deposition is larger than S deposition at most of the Intensive Monitoring plots in Western Europe, whereas the reverse is true for Central Europe. Total N inputs exceed a deposition level of 1000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> at approximately 45% of the plots. At those plots, adverse impacts on species diversity of ground vegetation are likely.
- Total deposition of acidity, caused by both S and N compounds varied mostly between 100 – 3000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. Very high inputs of acidity (> 3000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>) occur at approximately 15% of the plots, located in Western and Central Europe. Negative impacts on the forest ecosystem are likely at those plots
- Apart from the geographic region, atmospheric deposition is significantly influenced by altitude, tree height and rainfall.
- Concentrations of SO<sub>4</sub> and NO<sub>3</sub>, which are mainly influenced by S and N deposition, are significantly related to Al concentrations in acid soils. Molar ratios of Al to base cations above critical levels (levels indicative for adverse effects on roots) occur at some 10 – 20% of the plots.
- The variation in concentrations of major ions in the soil solution could to a large extent be explained by differences in atmospheric deposition and to a lesser extent by variations in meteorological conditions (specifically precipitation) and soil chemistry (especially pH or base saturation in relation to Al and to a lesser extent base cations)

Keywords: Intensive monitoring, data management, forest, crown condition, increment, meteorological stress, atmospheric deposition, soil solution chemistry

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## Preface

In order to gain a better understanding of the effects of air pollution and other stress factors on forests, a Pan-European Programme for Intensive and Continuous Monitoring of Forest Ecosystems has been implemented (the so-called level II programme). In this context 861 permanent observation plots for Intensive Monitoring of forest ecosystems have now been selected (512 in the European Union and 351 in several non-EU countries). The Pan-European Programme is based on both, the European Scheme on the Protection of Forests against Atmospheric Pollution (Council Regulation (EEC) No 3528/86) and the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests) under the Convention of Long-Range Transboundary Air Pollution (UN/ECE). The establishment of the Pan-European Programme was supported by Resolution No 1 of the first Ministerial Conference on the Protection of Forests in Europe (Strasbourg, 1990) and Resolution No 4 of the second Ministerial Conference on the Protection of Forests in Europe (Helsinki, 1993).

The Intensive Monitoring Programme aims at the assessment of crown condition, increment and the chemical composition of foliage and soil on all plots over a period of at least 15 to 20 years. Additional measurements foreseen on a limited number of plots include atmospheric deposition, meteorological parameters, soil solution chemistry and ground vegetation. In all these surveys, a number of mandatory and optional parameters has been defined. Additional studies, that are neither assigned as being mandatory or optional according to the relevant Regulation and Manual of ICP Forests, are also carried out in many countries, such as studies on phenology, phytopathology, and litterfall.

In order to set up procedures for the validation, storage, distribution and evaluation of the data at European level, a Forest Intensive Monitoring Co-ordinating Institute (FIMCI) has been set up being a contractor of the European Commission (EC). Within FIMCI, the DLO Winand Staring Centre for Integrated Land Soil and Water Research (SC-DLO) and Oranjewoud International work together. Apart from the data management, FIMCI also acts as an information centre for National Focal Centres (NFC's), including both EU-member states and non-member states.

By the end of 1996 the results of the four core surveys on the Intensive Monitoring plots (crown condition, soil, foliage, growth) and of atmospheric deposition were for the first time submitted to FIMCI. In 1997 these data and the data accompanying reports were validated and the first evaluations were presented in the second Technical Report of 1998. By the end of 1997, the NFC's submitted data for the second time, including first information on soil solution and meteo. These data were validated in 1998. The data presented in this years report includes the results of the annual surveys (crown condition, deposition, meteo and soil solution) for 1996. Furthermore it includes a further evaluation of the forest growth data of 1995 and 1996 that have partly been submitted earlier.

The target groups of this report are the active participants of the Intensive Monitoring Programme (National Focal Centres, National Involved Research Institutes, Scientific Advisory Groups, the Expert Panel Members, the Standing Forestry Committee of the European Union and ICP Forests) and the Scientific Community. The preparation of this report was possible thanks to the submission of data and information by the NFC's to FIMCI and the active participation and co-operation of the members and deputy members of the Scientific Advisory Group.



## **Extended Summary**

### ***The monitoring programme***

The Pan-European Intensive Monitoring Programme of Forest Ecosystems was started in 1994. The general aim of the Intensive Monitoring Programme is to contribute to a better understanding of the impact of air pollution and other factors which may influence forest ecosystems. At present, the programme covers 861 selected plots in 30 participating countries (512 plots in the EU and 349 plots in non-EU countries). Some surveys are carried out on all plots (crown condition, soil, foliage and forest growth). Furthermore, assessments of atmospheric deposition (505 plots), meteorology (180 plots), soil solution (238 plots), ground vegetation (618 plots) and remote sensing (approximately 150 plots) are executed at part of those plots. In total 774 Intensive Monitoring plots have already been installed. For most of the plots (around 85%) information on the methods applied has been received, validated and stored. The results presented in this report only include validated and stored data from 1996 or earlier with respect to crown condition (694 plots), forest growth (405 plots), atmospheric deposition (320 plots), meteorology (51 plots) and soil solution (103 plots). Soil and foliar data were described extensively in the previous Technical Report. Due to its non-systematic character the intensive monitoring data set is not representative for Europe in the statistical sense, but it does give information on stress and effects on a European-wide scale.

### ***Objectives***

The major aim of this year's report is to gain more insight in (i) the geographic variation of the atmospheric input of N, S and base cations (acidity) and (ii) the relationship between atmospheric deposition and soil solution chemistry, in view of differences in stand/site characteristics, meteorological conditions and soil chemistry. The latter relationship forms the basis for the assessment of input-output budgets, which in turn are a prerequisite to derive critical loads for a forest ecosystem. Furthermore, results of the increment or forest growth survey are further interpreted in view of their possible impact on atmospheric inputs. Unlike the first and second technical report, this report does not include a data evaluation strategy any more. Such a strategy is further updated and described in a separate document.

### ***Reliability and comparability of the data***

The reliability of the plot averaged data depends strongly on the number of observations or samples within the plot. A comparison of the calculated numbers that are required to derive a reliable plot-mean value and the actual numbers used, shows that both ranges coincide quite well for most of the surveys. The number of deposition samplers was nearly always equal or higher than the required minimum of 10. In case of soil solution, however, the numbers were comparatively small.

The reliability of data at European level is also influenced by the differences in data assessment methods. This aspect has specifically been investigated with respect to the deposition and soil solution surveys. In both cases, methods seem quite comparable at most of the plots. For monitoring of throughfall data, use was mostly made of funnels. Gutters were used at a minority of the plots. With respect to the analyses of the water samples, results of a ringtest, which involved 18 laboratories participating in the Intensive Monitoring programme, showed no

comparability problems for the concentrations of major ions in the bulk precipitation and throughfall (5.9% outliers).

For monitoring of the soil solution, use was made of suction cups (sometimes combined with zero tension lysimetry) at the majority (89%) of the plots. Furthermore, the majority of used lysimeters is made of materials that are considered appropriate, such that the sample solution is not influenced by the sampler itself. The ion concentrations obtained by non-destructive methods (zero tension lysimeters or suction cups) do differ from those obtained by destructive methods (centrifugation and the saturation extract method) used at the remaining plots, since different types of soil water are extracted. In general, concentrations increase going from zero tension lysimeters to suction cups and to centrifugation. Some information on those differences is given in an Annex, based on a comparative study in Finland and the Netherlands. An in-depth study on the differences is foreseen by Finland and Denmark. It is expected that results will become available this year and that a summary can be included in next years report.

### *Data quality assurance*

Data consistency checks were carried out on all the data submitted in the various surveys. This includes on the validity of codes and the plausibility of results of parameters or parameter combinations. Special attention was given to the quality assurance and quality control (QA/QC) of the chemical composition of bulk deposition, throughfall, stemflow and soil solution. This included a check on:

- the balance between cations and anions
- the difference between measured and calculated electric conductivity
- the ratio between Na and Cl concentrations.

Checks on the ionic balance and the difference between measured and calculated electric conductivity were only made when all cations and anions were measured. The only allowances made were that Al was missing at a pH > 5 and alkalinity at a pH < 5 and that dissolved organic carbon (DOC) was missing. For soil solution, checks on the ionic balance could only be made for a very limited part of the measurements, mainly due to neglect of Na and Cl.

Results for the balance between cations and anions in bulk deposition, throughfall and stemflow showed large differences varying from a strong anion excess to a large cation excess. Approximately 50-60% of the measurements only appeared to fulfil the requirement that the percentage difference is less than 20%. Results were better for the soil solution samples. The percentage of measurements in an acceptable range of  $\pm 20\%$  was nearly 90% when DOC was taken into account, but it decreased to less than 60% when DOC was neglected. In that case, there was generally a large cation excess. This implies that a reliable charge balance check can only be made when all major cations and anions, including DOC, are available. Results for the difference between measured and calculated electric conductivity were slightly better than the difference between the sum of cations and anions. An allowable discrepancy between measured and calculated conductivity of 20% was fulfilled by approximately 55% of the measurements for bulk deposition, 65% for throughfall, 75% for stemflow and 85% for the soil solution.

On average the Na/Cl ratio in bulk deposition, throughfall, stemflow and soil solution resembled those in seawater ( $0.858 \text{ eq.eq}^{-1}$ ), but a high variation occurred, especially in bulk deposition. The resemblance with seawater was better at higher concentrations. Especially at low Cl concentrations, quite a lot of extremely high Na/Cl ratios were found, exceeding values of 10 up

to 100. Those ratios were also correlated was a consistent cation excess, thus indicating Na contamination. There are clear indications that solution samples that are kept in the field in ordinary glass bottles are contaminated by Na release from the bottle.

The results of the various QA/QC procedures on the chemical composition of deposition and soil solution samples poses questions to the quality of the deposition data. Because of these questions, interpretations of atmospheric deposition have been carefully considered in view of its plausibility in the light of available literature.

### ***Crown condition***

Crown condition, in terms of defoliation and discoloration, is assessed at all Intensive Monitoring plots. The evaluation of crown condition data focused on changes in defoliation, while checking the comparability of data assessment methods between 1996 and 1995. Elevated discoloration values were observed at a limited number of plots (<10%) and differences between 1996 and 1995 were small. An in-depth interpretation was not yet carried out, since this is hampered by a lack of information on stand history, pests and diseases, meteorological stress and air quality at most of the plots.

Results for defoliation showed a more or less significant deterioration in crown condition for European Oak, Pine, Spruce and Fir, whereas a significant recovery was observed for Alpine Conifers, Mediterranean Conifers and Evergreen Oak. Data assessment methods generally appeared to be comparable between 1995 and 1996. However, at approximately 25% of the plots either the assessment dates appeared to differ more than one month or the number of trees that were assessed in 1996 differed from 1995, in 20% of the cases being larger in 1996.

### ***Forest growth***

The evaluation of forest growth data was limited to 403 plots, for which data and data assessment methods were received and stored. Data on both tree diameter (at breast height) and tree height at tree level were used to calculate a number of indices related to site quality, stand density, stand structure and (tree) species diversity, that influence the input of atmospheric pollutants by differences in the scavenging of gases and aerosols (dry deposition). Furthermore, it may influence evapotranspiration and thereby soil solution chemistry.

The available information on the measurements of stem diameters and tree heights offered good possibilities to derive relevant stand characteristics for about 65% of the approximately 400 Intensive Monitoring Plots. At approximately one-third of the plots, data evaluation was limited by incomplete measurements (not all trees in a plot or subplot were assessed) or lack of methodological information. This aspect requires improvement in new assessments (submissions). The evaluated stands showed a considerable variation in stand density, stand structure and stand height. Approximately 40% of the stands were complete monocultures, whereas the remaining stands were mostly slightly mixed, except for many Oak stands. Most stands were relatively even-aged, except for the Evergreen oak stands.

### ***Atmospheric deposition***

The total number of plots at which deposition measurements took place in 1996 equalled approximately 300 plots in a total of 18 countries. With the exception of Poland, with a total

number of 122 bulk deposition plots, throughfall was measured at all those plots. Stemflow was only measured at 23 plots in 8 different countries. The number of plots where the number of measurements was such that annual deposition fluxes could be calculated was slightly lower. Major conclusions related to annual atmospheric deposition data at those Intensive Monitoring plots aspects are given below.

*The relative contribution of wet and dry deposition in the potential acid input*

On average, the contribution of dry deposition was at least one-third of the total deposition for both the S and N compounds, whereas it was slightly lower for base cations. The contribution of wet deposition was dominant in Northern Europe, whereas dry deposition was relatively important in parts of Western and Central Europe.

*The relative contribution of N and S compounds and of base cations in the atmospheric input*

On average, the N input in bulk deposition and throughfall equalled the S deposition. The average calculated total N deposition was, however, 50% larger than of S. Even though this result may be influenced by the calculated N uptake rates, it implies that N is a dominating factor in the acidic input in large parts of Europe. The relative contribution of  $\text{NH}_4$  and  $\text{NO}_3$  in N deposition varied largely over the plots but in most countries, especially in Northern and Central Europe,  $\text{NH}_4$  seems the dominating N compound at most of the plots. A significant relationship was further observed between the input of Ca and  $\text{SO}_4$  both in bulk and total deposition. The correlation may partly be due to associated emissions of  $\text{SO}_2$  and Ca from smelters and refineries.

*Ranges and geographic variation of atmospheric inputs in view of critical loads*

Total deposition of S and N compounds ranged between 100-3000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  (approximately 2-45  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ) at approximate 90% of the plots, but values up to 4000-8000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  were also observed for N and S, respectively. Both bulk and total deposition of N generally appeared to be higher than S deposition at plots in Western Europe (UK, Belgium, Netherlands, Luxembourg, France), whereas the opposite was generally observed at plots in Central Europe (Poland, Czech Republic, Austria, Hungary). For the base cations, the range was mostly between 100 and 2000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  for the sum of Ca, Mg and K. The geographic pattern of bulk deposition of Ca, being the most important base cation neutralising the acid input from the atmosphere, is low in Northern Europe, high in Central and Southern Europe and intermediate in Western Europe. In Central and Southern Europe the acidic input is thus largely set off by the input of base cations.

Approximately 45% of the considered plots received an N input above 1000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  (14  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ), being a deposition level at which the species diversity of the ground vegetation may be at risk. Below this deposition level, tree growth may, however, be inhibited. Critical loads related to tree health are higher and deposition levels vary from approximately 1000-3500  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ . A comparison with present loads shows that those impacts most likely occur at several plots. The total input of acidity, being the input of S and N compounds minus the deposition of accompanying base cations corrected for Cl, ranged mostly between 200-4000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ . Considering a variation of critical acid loads related ratios of approximately 1500-3500  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ , impacts are likely at part of the plots. A specific comparison of present and critical acid loads is needed to assess the risk of the acid atmospheric input in terms of elevated  $\text{Al}/(\text{Ca}+\text{Mg}+\text{K})$  ratios, affecting root growth and root uptake.

*Relationships between atmospheric deposition and environmental factors*

Literature data indicate a clear impact of site characteristics such as (climatic) region, altitude, and stand characteristics, such as tree species, stand structure and stand height, on atmospheric

deposition. A first investigation was thus performed of the relationships between the key parameters in both surveys and those available stand and site characteristics. Geographic region appears to have a dominant influence on a limited data set of deposition data in combination with the various environmental factors. This follows from the results of both a principal component analyses and a multiple regression analyses. Atmospheric deposition of all ions is significantly lower in the Boreal regions compared to Western Europe, whereas SO<sub>4</sub>, NO<sub>3</sub> and Ca deposition is significantly higher in the Central/Eastern part of Europe. There is furthermore a highly significant positive correlation of atmospheric deposition and rainfall, except for NH<sub>4</sub> and K. The deposition of S and N compounds appears to decrease significantly with an increase in altitude. For base cation deposition, the impact of altitude is, however, insignificant.

### ***Meteorological data***

Meteorological data were available at 51 plots in seven countries. Those data did allow the calculation of key parameters in view of their possible impact on crown condition and forest growth, such as to low temperatures (e.g. late frost) and high temperatures (e.g. summer index at 35 plots) and drought stress (e.g. relative transpiration at 12 plots). Late frost occurred on 25 of the 35 plots. Lowest summer indices (temperatures) were of course found in Northern Europe (Finland, Denmark and the northern part of the UK) and at high altitude plots in France.

Drought stress in terms of computed transpiration reductions (reduction of potential evapotranspiration) varied between 34% to about 60% for 12 intensive monitoring plots, located in France, Luxembourg and Greece. The quality of the calculated drought stress indices depends on the quality of the model and the data. Validation was only possible for the interception module of the hydrological model. This was done by comparing computed interception and measured interception (rainfall minus throughfall) at 8 of the 12 plots, where all data were available. Results showed that the actual and simulated interception compare reasonably well for 6 of the 8 plots. An indication of the quality of the precipitation data was obtained by evaluating the consistency of the sum of daily precipitation from the meteorological data set and the (bulk) deposition data at 20 plots. For 70% of the plots the deviation was less than 5%; only occasionally 10% deviation was found. This means that yearly precipitation sums seem quite reliable. Nevertheless, the computations performed with the 1996 meteorological data only have a tentative character, since the available data set was very limited and the simulation of drought stress could only partly be validated.

### ***Soil solution chemistry***

Data for the soil solution chemistry in 1996 were stored for a total of 103 plots in eight countries concentrated in Western and Northern Europe. The evaluation focused on the chemistry of major ions in soil solution impacted by N and S deposition, either directly (SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>) or indirectly through soil buffering reactions (H, Al, Ca, Mg, K). Major conclusions are given below.

#### ***Relationships between element concentrations in soil solution***

The concentration of potentially toxic Al in both topsoil and subsoil was strongly related to the concentration of SO<sub>4</sub> and NO<sub>3</sub> in acid soils (soils with a base saturation below 25% or a pH below 4.5). In those soils, the acid input is mainly neutralised by release of Al. Above those base saturation and pH levels, the relationship between the concentration of Al versus SO<sub>4</sub> plus NO<sub>3</sub> was very weak, indicating that the acidity is neutralised by the release of base cations. The impact of base saturation on the relationship between Ca and strong acid anions was less. The (logarithmic) concentration of the calculated free Al activity, which is most toxic to roots, was

strongly related to the pH. The results indicate that Al release is dominated by complexation with soil organic matter, especially in the organic layer.

#### *Range in element concentrations in view of critical levels*

Concentrations of  $\text{SO}_4$ ,  $\text{NO}_3$ , total N, Al and Ca was lower than  $2000 \text{ mmol}_c.\text{m}^{-3}$ , whereas concentrations of  $\text{NH}_4$  were nearly always lower than  $1000 \text{ mmol}_c.\text{m}^{-3}$ . The concentrations of  $\text{NO}_3$  in soil solution exceeded the official ground water quality criterium of  $800 \text{ mmol}_c.\text{m}^{-3}$  at 10-15% of the plots, depending upon the depth considered. The ground water quality criterium of  $20 \text{ mmol}_c.\text{m}^{-3}$  for Al was exceeded at 60% of the plots at greater depth. Note, however, that concentrations in soil solution are generally higher than in ground water. The  $\text{Al}/(\text{Ca}+\text{Mg}+\text{K})$  ratios exceeded a critical ratio of 1.0 in approximately 10-15% of the plots, depending on the layer considered. This ratio can be considered indicative for negative impacts on tree roots. Both the  $\text{NH}_4/\text{K}$  ratio and  $\text{NH}_4/\text{Mg}$  ratio hardly ever exceeded a critical value of 5.0 in the mineral soil.

#### *The simultaneous impact of atmospheric deposition, meteorological conditions and soil chemistry on the soil solution chemistry.*

The variation in concentrations of major ions in the soil solution could to a large extent be explained by differences in atmospheric deposition and to a lesser extent by variations in meteorological conditions (specifically precipitation) and soil chemistry (C/N ratio, pH or base saturation whenever relevant). With the exception of  $\text{SO}_4$ , the deposition of  $\text{NH}_4$  had a (highly) significant impact on all the considered compounds, increasing the concentration of N compounds, base cations and Al and decreasing the pH. This can be explained by the acidifying impact of  $\text{NH}_4$  deposition, caused by the conversion of  $\text{NH}_4$  to  $\text{NO}_3$  (nitrification) in the soil and the subsequent release of base cations and Al buffering the acid input. The impact of the deposition of  $\text{SO}_4+\text{NO}_3$  on base cations and Al was generally slightly lower. This result should, however, be interpreted with care, since there is a high correlation between the deposition of  $\text{NH}_4$ ,  $\text{NO}_3$  and  $\text{SO}_4$ .

Using the original data, the explained variation in element concentrations by environmental factors mostly ranged between 45 and 75%. In nearly all situations, atmospheric deposition of the considered compound was the most important influencing factor with the exception of  $\text{NO}_3$ .  $\text{NO}_3$  deposition had no significant impact on the concentration of  $\text{NO}_3$  in both the topsoil and subsoil, whereas  $\text{NH}_4$  deposition was highly significant in both cases. In general,  $\text{NH}_4$  deposition was an equally good predictor for the concentration of Ca, Mg and K as the throughfall of the cation itself. The precipitation or the precipitation excess mostly had a significant to highly significant effect on the concentrations of major ions, especially when using the log-transformed data. In most cases base saturation did not have a significant influence on the Al and base cation concentrations. It did, however strongly affect the pH. The influence of the C/N ratio on the measured  $\text{NO}_3$  and  $\text{NH}_4$  concentration was mostly negligible.

#### ***Final conclusions and discussion***

Focusing on the results of atmospheric deposition and soil solution chemistry, the following main conclusions have been drawn:

- Total nitrogen input is larger than sulphur input at most of the Intensive Monitoring plots in Western Europe, whereas the reverse is true for Central Europe. Total deposition of both compounds was clearly correlated and varied mostly between  $100 - 3000 \text{ mol}_c.\text{ha}^{-1}.\text{yr}^{-1}$ . On average, N deposition was, however, 50% larger than S deposition, exceeding a deposition level of  $1000 \text{ mol}_c.\text{ha}^{-1}.\text{yr}^{-1}$  ( $14 \text{ kg}.\text{ha}^{-1}.\text{yr}^{-1}$ ) at approximately 45% of the plots. At those plots,

adverse impacts on species diversity of ground vegetation are likely. Impacts of the input of acidity, caused by both S and N compounds are likely at very high deposition levels ( $> 3000 \text{ mol}_c.\text{ha}^{-1}.\text{yr}^{-1}$ ) that occur at approximately 15% of the plots, located in Western and Central Europe. The negative impacts are partly set off by base cation deposition, especially in Central (and Southern) Europe. The impact of geographic region can be biased by the uneven representation of plots in those regions.

- Deposition of  $\text{SO}_4$  and N strongly influences the concentrations of  $\text{SO}_4$  and  $\text{NO}_3$ . Concentrations of those ions are significantly related to the Al concentration in acid soils (soils with a  $\text{pH} < 4.5$  or a base saturation  $< 25\%$ ), especially in the subsoil. In slightly acid to near neutral soils, Ca concentrations are significantly related to  $\text{SO}_4$  and  $\text{NO}_3$ . pH was a highly significant predictor for the activity of free (uncomplexed) Al that is most toxic to roots. Concentrations of  $\text{NO}_3$  and ratios of Al to Ca+Mg+K above levels, that are indicative for adverse effects, occurred at some 10 – 20% of the plots. Those percentages should be considered indicative only, because they are either based on (i) ground water quality criteria ( $\text{NO}_3$ ), deviating from soil solution or (ii) laboratory based experiments that may deviate from the actual field situation (Al/(Ca+Mg+K) ratios).
- Apart from the geographic region, atmospheric deposition is significantly influenced by altitude, tree height and rainfall. The variation in concentrations of major ions in the soil solution could to a large extent be explained by differences in atmospheric deposition and to a lesser extent by variations in meteorological conditions (specifically precipitation) and soil chemistry (especially pH or base saturation in relation to Al and to a lesser extent base cations). These kind of relationships may be used for upscaling of the results to e.g. level 1 plots when (i) the relationships largely explain the variation in response variable and (ii) the predictor variables are available at level 1 plots or can be estimated with reasonable accuracy.



# 1 Introduction

In order to gain a better understanding of the effects of air pollution and other stress factors on forest ecosystems, a Pan-European Programme for Intensive and Continuous Monitoring of Forest Ecosystems has been implemented. This chapter first presents information on the background and current status of the Intensive Monitoring Programme (Section 1.1). It then highlights the focus of this year's Technical Report in view of the overall objectives of the programme (Section 1.2) and it ends with a description of the content of the Technical Report (Section 1.3). The target groups of this report are the active participants of the Intensive Monitoring Programme (National Focal Centres, National Involved Research Institutes, Scientific Advisory Groups, the Expert Panel Members, the Standing Forestry Committee of the European Union and ICP Forests) and the Scientific Community.

## 1.1 Background and current status of the Intensive Monitoring Programme

Based on the agreed selection criteria, laid down in Commission Regulation (EC) N° 1091/94, the EU Member States started to select and install their plots in 1994. After acceptance of the relevant parts of the ICP Forests Manual (Task Force meetings in Lillehammer and Prague, 1994 and 1995), also the non-EU countries started with the selection and installation process. In January 1995 and January 1996 progress reports on the selection and installation of the Intensive Monitoring plots were prepared (e.g. EC, 1996; 'orange brochure'). Since then several countries have reviewed their selection and in some cases amendments have been made. In 1997, a first technical report on the results of the 'Pan-European Programme for the Intensive Monitoring of Forest Ecosystems' in Europe has been published including information on: (i) the implementation of the programme, (ii) the procedures for the management of the data and information described in data accompanying reports (DAR's), (iii) the contents of the database and DAR's obtained so far for crown condition, soil, foliage, increment and deposition and (iv) a first set-up of an evaluation strategy for the data.

At present, a total of 861 observation plots have been selected for this second level of monitoring intensity. In the European Union 512 permanent observation plots for Intensive Monitoring of forest ecosystems have been selected and installed. In several non-EU countries, including Belarus, Bulgaria, Croatia, Czech Republic, Estonia, Hungary, Latvia, Lithuania, Norway, Poland, Romania, Russia (St. Petersburg-region), Slovak Republic, Slovenia and Switzerland, 351 plots have been selected whereas 250 plots have been installed.

The Intensive Monitoring Programme contains the assessment of crown condition, forest growth (increment) and the chemical composition of foliage and soil on all plots. Additional measurements on a limited number (at least 10%) of the plots include atmospheric deposition, meteorological parameters, soil solution chemistry and ground vegetation. In the expert panels on deposition and meteorology, the possible inclusion of ambient air quality ( $O_3$ ,  $SO_x$ ,  $NO_x$  and  $NH_x$ ) and of phenology are discussed. Within each of these surveys, a number of mandatory and optional parameters have been defined. The temporal resolution of the surveys is scheduled as follows:

- crown condition (at least once a year)
- chemical composition of the contents of needles and leaves (at least every 2 years)
- soil chemistry (every 10 years)

- increment / forest growth (every 5 years)
- atmospheric deposition (continuous)
- soil solution chemistry (continuous)
- meteorology (continuous)
- ground vegetation (every 5 years)
- remote sensing/aerial photography (once)

## **1.2 Aim of the report**

### ***Overall aim***

The Technical Reports on the ‘Pan-European Programme for the Intensive Monitoring of Forest Ecosystems’ in Europe differ each year in contents in view of the increased data availability in time. The first report, presented in 1997 only included a description of the contents of the database and of the data assessment methods for the core surveys and a data evaluation strategy. The second technical report presented in 1998, focused strongly on a description of data assessments (data coverage, data comparability, data reliability) and on (preliminary) results of key parameters in the five core surveys (crown condition, soil, foliar composition, forest growth and atmospheric deposition). Even though that report included preliminary correlative studies between key parameters and major stand and site characteristics, it did not contain in-depth evaluations of the data in several surveys, since the data did not yet allow such evaluations. Instead, it included examples of data evaluations from other research programs with (comparable) data. Those assessed in the ‘Intensive Monitoring Programme’, to illustrate the potential use of the data set.

An important aim of this year’s report is to gain more insight in atmospheric deposition and its relationship with soil solution chemistry. This relationship forms the basis for the assessment of input-output budgets, which in turn are a prerequisite to derive critical loads for a forest ecosystem. The focus (evaluations and key parameters included) is described more specifically below.

### ***Data evaluation in view of the overall objectives of the Programme***

The major aim of the ‘Pan-European Programme for the Intensive Monitoring of Forest Ecosystems’ is to gain a better insight in the impacts of air pollution (specifically the elevated deposition levels of SO<sub>x</sub>, NO<sub>x</sub>, NH<sub>x</sub> and O<sub>3</sub>) and other stress factors on forest ecosystems. Scientific evaluations should thus be focused on the relationships between the parameters describing the forest condition (such as defoliation, growth and nutrition) and the influencing parameters (such as site and stand characteristics, meteorology and deposition).

For a large number of monitoring plots in Europe (approximately 300-850 depending on the data considered), the Intensive Monitoring database will ultimately contain data on:

- Site factors: stand and site characteristics, stand history/management
- Stress factors: meteorological data and air pollution / atmospheric deposition data, pests and diseases
- ‘Biological’ ecosystem condition: crown condition, forest growth, species diversity of the ground vegetation
- ‘Chemical’ ecosystem condition: foliar chemistry, soil chemistry, soil solution chemistry

In terms of data availability, the Intensive Monitoring database thus allows to derive relationships between (trends in) stress (site and stress factors) and (trends in) effects (chemical and ecological ecosystem condition), being the most important aim of the Intensive Monitoring Programme. An overview of the most relevant relationships between the ultimately available data in the Intensive Monitoring database is given in Fig. 1.1.

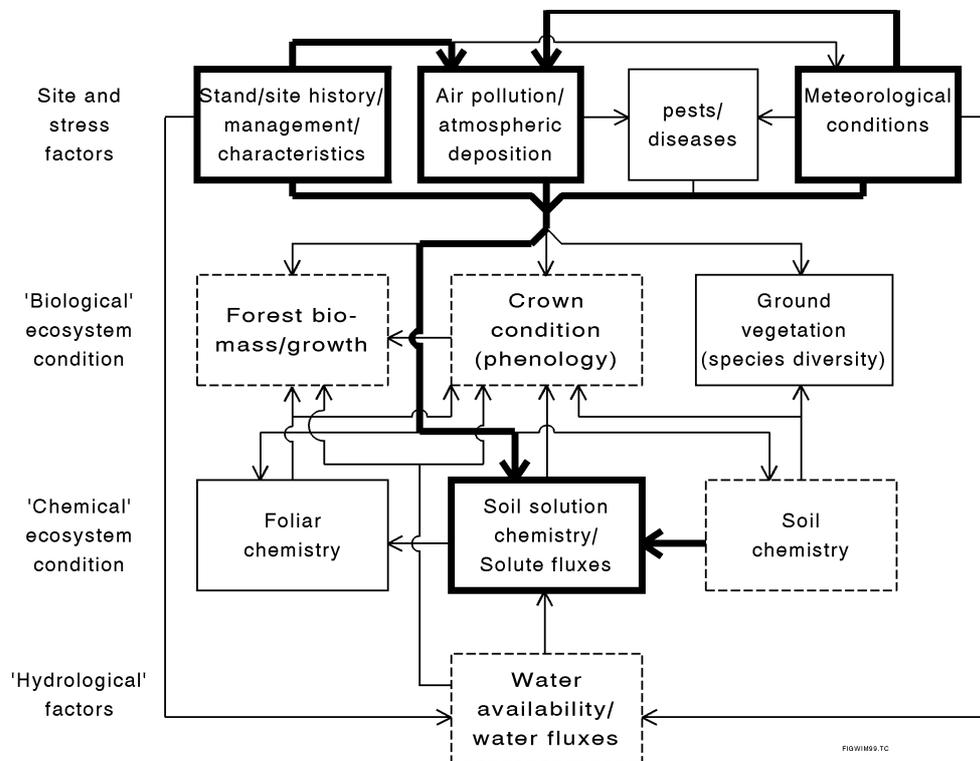


Figure 1.1 Flow diagram illustrating the relationships between site and stress factors and the forest ecosystem condition. Boxes and arrows in bold are specifically investigated in this year's report. Attention is further given to Boxes with dotted lines.

This year's report focuses on atmospheric deposition, meteorology and soil solution chemistry. Methods and results are specifically focused on (i) the relationship between atmospheric deposition and stand/site characteristics and meteorological conditions and (ii) the simultaneous impact of atmospheric deposition, meteorological conditions and soil chemistry on the soil solution chemistry (Boxes and arrows in bold in Figure 1.2).

Unlike the first and second technical report, this report does not include a data evaluation strategy any more. An updated evaluation strategy, including a time frame for the next 15 years, is described in a separate document (De Vries, 1999).

### **Selection and presentation of key parameters**

An overview of all relevant key parameters in the various surveys is given in Table 1.1. The key parameters considered in this year's report are given in bold. More information on the background of these key parameters is given in Klap et al (1997) and in De Vries et al (1998).

Table 1.1 Key parameters describing the available 'ecological and chemical' forest condition and stress. Values in bold are included in this year's report.

Type of parameter	Key parameter	
Ecological condition	<ul style="list-style-type: none"> <li>- <b>crown condition</b></li> <li>- <b>increment</b></li> <li>- ground vegetation</li> </ul>	<p><b>Defoliation</b>, discoloration  <b>Diameter, tree height, Stand Density Index<sup>1)</sup>, Site structure Index<sup>1)</sup></b>  ground vegetation index</p>
Chemical condition	<ul style="list-style-type: none"> <li>- foliar composition</li> <li>- soil composition <ul style="list-style-type: none"> <li>• carbon</li> <li>• nutrients:</li> <li>• acidity:</li> <li>• toxic elements:</li> </ul> </li> <li>- <b>soil solution chemistry</b></li> </ul>	<p>N, P, S, Ca, Mg, K, N/P, N/Mg, N/K, Fe, Mn, Cu, Zn  C  N, P, Ca<sup>2)</sup>, Mg<sup>2)</sup>, K<sup>2)</sup>, C/N, N/P  pH, base saturation<sup>3)</sup>  Pb, Cd, Cu, Zn  <b>SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, Ca, Mg, K, Al, pH, DOC</b></p>
Stress	<ul style="list-style-type: none"> <li>- stand and site characteristics</li> <li>- biotic stress</li> <li>- air pollution</li> <li>- <b>atmospheric deposition</b></li> <li>- <b>meteorology</b></li> </ul>	<p>Tree species, tree age, climatic region, altitude, soil type  Easily assessable damage types  O<sub>3</sub>, SO<sub>2</sub>, NO<sub>x</sub> and NH<sub>3</sub> concentrations in air  <b>SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, Ca, Mg, K, pH in bulk deposition, throughfall and stemflow</b>  <b>precipitation, temperature, evapotranspiration<sup>1)</sup></b></p>

<sup>1)</sup> Those parameters are derived from basic data.

<sup>2)</sup> Limited to the organic layer only.

<sup>3)</sup> Limited to the mineral layer only.

### 1.3 Contents of the report

Chapter 2 provides information on the current implementation (plot selection and data submission) of the Intensive Monitoring Programme. General methodological aspects, such as the reliability/comparability and the statistical evaluation of data are described in Chapter 3. The Chapters 4 to 8 present the methodological approaches and the results of evaluations related to key parameters in the surveys on crown condition (Chapter 4), forest growth (Chapter 5), atmospheric deposition (Chapter 6), meteorological parameters (Chapter 7) and soil solution chemistry (Chapter 8). The methodological approach refers to methods for data assessment, data quality assurance (if applicable) and data evaluation.

Results of the crown condition survey are limited to a comparison of 1996 and 1995 (Chapter 4). The data in the forest growth survey are only evaluated in terms of stand characteristics influencing the atmospheric inputs, such as dominant height of the stand, stand structure index and stand density index (Chapter 5).

Results of the survey on atmospheric deposition focus on relationships between (i) bulk deposition, throughfall and stemflow to gain insight in total atmospheric deposition, accounting for canopy interaction fluxes and (ii) throughfall or calculated total atmospheric deposition and stand/site characteristics, including those derived from the forest growth survey (Chapter 6). Meteorological data are evaluated in terms of indicators for temperature stress, such as the (late) frost and summer index, and water stress (relative transpiration). Considering the limited amount of information, these data are not used in further evaluations (Chapter 7).

Data on element concentrations in the soil solution are evaluated in view of (i) correlations between the acid-base chemistry of the soil solution and the sulphate and nitrogen concentrations, (ii) possible exceedance of critical levels and (iii) overall combined influence of stand and site

characteristics, meteorology, soil chemistry and atmospheric deposition on the soil solution chemistry (Chapter 8). Chapter 9 contains the discussion and several conclusions related to the results presented in Chapter 4 to 8.



## **2 The Intensive Monitoring Programme: plot selection and data submission**

In contrast to the monitoring at the systematic (16x16km) grid that comprises crown condition and the chemical status of soil and foliage, the Intensive Monitoring Programme is carried out on selected plots, and also comprises monitoring of increment, deposition, meteorology, soil solution and ground vegetation. Due to its non-systematic character, the intensive monitoring data set is not representative for Europe in the statistical sense, although it does give information on stress and effects on a European-wide scale. In this chapter an overview of selected plots in the various surveys (Section 2.1) and of the data that have been stored until 1996 (Section 2.2) are presented.

### **2.1 Selected plots in the various surveys**

The Intensive Monitoring Programme now includes 861 selected plots. The selection of the plots is completed in 30 participating countries. Some countries that participate in the ICP Forests programme, have indicated their participation in the Intensive Monitoring programme, but have not sent the general plot information yet. For some other European countries, it is not yet sure whether and when they intend to join the Intensive Monitoring Programme. With the possible inclusion of these countries the total number of plots could rise to approximately 900.

Table 2.1 shows the number of plots selected and installed and the number of plots on which the different surveys (crown condition, soil, foliage, increment and deposition, soil solution, meteorology and ground vegetation) are (planned to be) executed. Four surveys have to be conducted on all plots (crown condition, soil, foliage and increment). Deposition, soil solution, meteorology and ground vegetation should be conducted on at least 10% of the plots, whereas aerial photographs is optional. According to the information received, atmospheric deposition is carried out at 505 plots. Surveys with respect to meteorology and soil solution are carried out at 180 and 238 plots respectively. Based on information submitted by the countries (which is not yet complete), it can be concluded that ground vegetation surveys will be carried out at 618 plots (Table 2.1), whereas the application of aerial photography is foreseen at approximately 150 plots. Several countries also plan to carry out additional surveys on the plots, such as phytopathology, litterfall, study of lichens and/or mosses, mycorrhiza and/or fungi and other in-depth studies to soil water regimes, gas exchange and intensive air quality measurements. The number of plots that have presently been installed equals 774.

An overview of the surveys carried out at the different plots is given in Fig. 2.1. Fig. 2.1 does not yet include ground vegetation, since we do not yet know the geographic location of all plots. This map is based on information submitted until February 1999. The map indicates that the (relative) number of plots at which the continuous surveys on deposition, meteorology and soil solution are carried out varies strongly between countries. It should be noted that for 1 plot in Greece, 2 plots in Italy and 1 plot in Hungary, increment measurements are not carried out, due to the strongly deviating vegetation structure at these plots.

Table 2.1 Overview of the number of selected plots for the main surveys<sup>1)</sup>

Countries	Total	Crown	Soil	Foliar	Increm.	Atm. Depo	Meteo	Soil sol.	Gr. Veget.
EU countries									
AU	20	20	20	20	20	20	2	2	20
BL <sub>v</sub>	12	12	12	12	12	6	2	6	12
BL <sub>w</sub>	8	8	8	8	8	2	2	2	8
DK	16	16	16	16	15	10	3	10	15
DL	88	88	88	88	88	86	51	54	71
EL	4	4	4	4	3	4	4	2	4
ES	53	53	53	53	53	11	11	6	53
FR	100	100	100	100	100	25	25	15	100
IR	15	15	15	15	15	3	8	3	9
IT	26 <sup>1)</sup>	24	20	19	20	16	9		19
LX	2	2	2	2	2	1	2		2
NL	14	14	14	14	14	4		14	14
PO <sub>m</sub>	9	9	9	9	9	1	1	1	9
PO <sub>açor</sub>	4	4	4	4	4	1	1	1	
SF	31	31	31	31	31	16	12	16	31
SW	100	100	100	100	100	50		50	
UK	10	10	10	10	10	10	2	7	10
Total EU	512	510	506	505	504	271	147	194	377
non-EU countries									
BG	3 <sup>2)</sup>	3	3	3	3	3	3	3	3
BY	81 <sup>2)</sup>	81	81	81	81				
CH	17 <sup>3)</sup>	17	17	17	17	13	17	7	17
CZ	10	10	10	10	10	4	1	1	10
EE	7	7	7	7	7	5	0	2	7
HR	7	7	7	7	7	2	3	3	4
HU	14	14	14	14	14	14	7		14
LT	9	9	9	9	9				9
LV	2	2	2	2	2	2	2	2	2
NO	17	17	17	17	17	17		17	17
PL	148	148	148	148	148	148			148
RO	13	13	13	8	13	4		4	13
RU	12	12	12	12	12	12			
SL	3 <sup>2)</sup>	3	3	3	3	2	3		
SR	7	7	7	7	7	7			
Total non-EU	350	350	350	345	350	234	33	44	241
Total	862	860	847	847	852	505	180	238	618

<sup>1)</sup> The numbers of installed plots in Italy is 24 (1998).

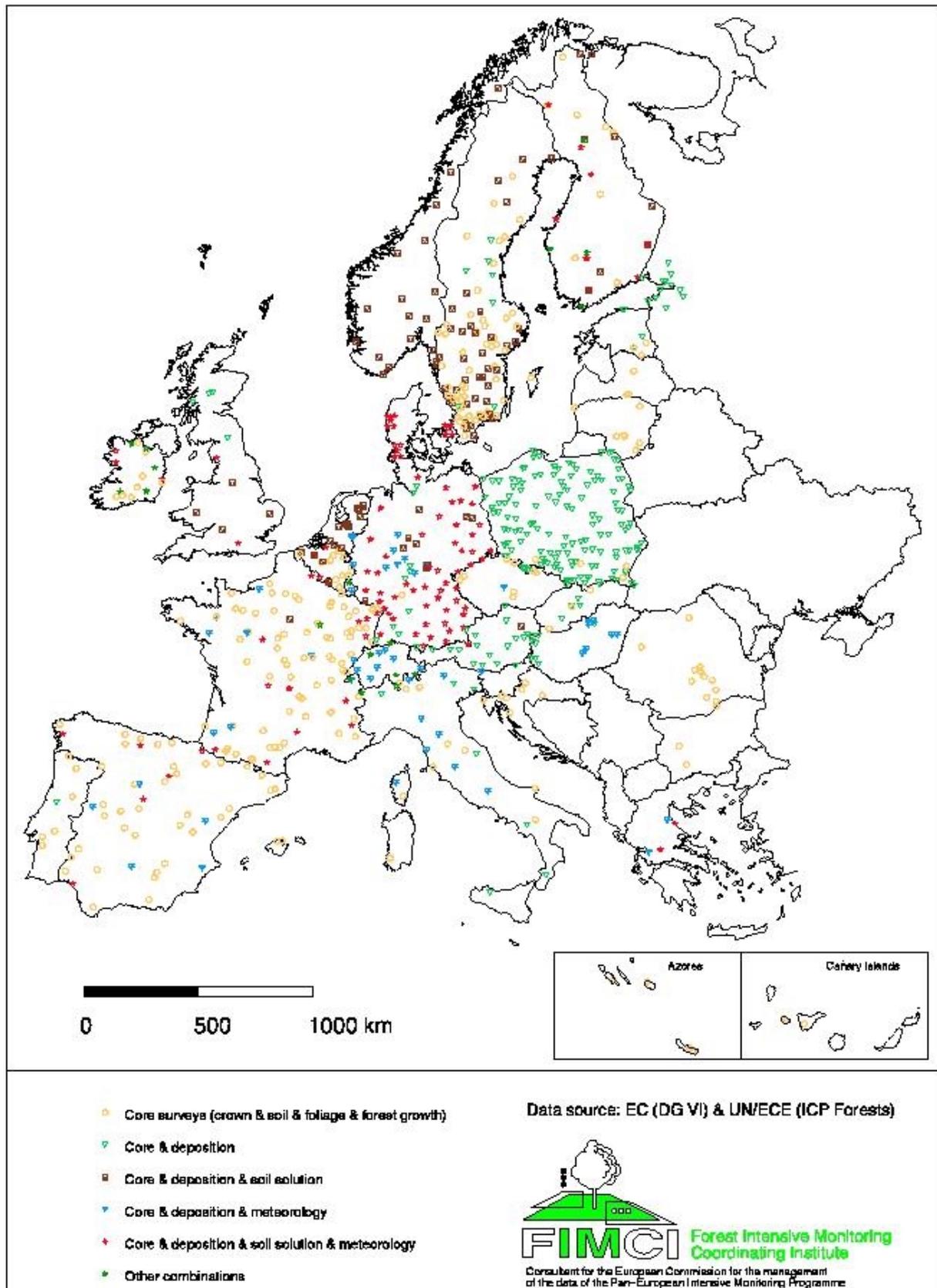
<sup>2)</sup> In these countries plots have not yet been installed.

<sup>3)</sup> The numbers of installed plots in Switzerland is 16

## 2.2 Submitted data and information until 1996

Table 2.2 gives an overview of the number of installed plots, and the number of plots for which data, DARQ and both data and DARQ's are stored. This table shows that the number of plots for which data were stored (and DAR-Q information is available) is substantially lower than the number of installed plots. The main reasons for this difference are:

- some countries have not submitted data for some of the surveys
- some countries submitted data that have still not been stored because the data are not yet complete or problems persist with respect to their quality
- at some of the installed plots, monitoring has started only very recently. Consequently, no data or DAR-Q information is available yet



**Figure 2.1 Geographical distribution and surveys carried out at the plots of the Pan-European Intensive Monitoring Programme (based on information received until April 1999)**

Compared to last years' report, the number of plots with data for 1996 has increased for most surveys. This indicates that also in 1998 data and corrections for the year 1996 were received. Nevertheless, for a number plots, problems with 1996 data remain that can hopefully be solved in the coming year. Furthermore table 2.2 shows that for the vast majority of the plots with stored data, also the DARQ information is available.

*Table 2.2 General overview of the submitted data and information for the seven surveys until the year 1996*

Survey	Number of installed plots <sup>1)</sup>	Number of plots with data stored (for 1996 <sup>2)</sup> )	Number of plots with DAR-Q information stored	Number of plots with both data en DAR-Q information
Crown condition	859	694	700	667
Soil condition	847	504	647	499
Foliar condition	847	494	520	483
Increment	852	405	440	345
Deposition	505	320	395	263
Meteorology	180	51	132	51
Soil solution	238	103	145	62

<sup>1)</sup> The number of plots for which plot characteristics were received, was less: 762 (see Section 2.1)

<sup>2)</sup> For soil, foliage and increment also data from earlier years have been used. If a country submitted data for more than one year, only the data from the most recent year were used in the evaluations.

## **3 General methodological aspects**

### **3.1 Data quality checks and data presentation**

#### *Data quality checks*

The procedures described in the ‘Strategy plan for the validation and evaluation of data’ (De Vries et al., 1996), were used as a guideline to streamline the data and information flows. However, changes were made to the designed procedures whenever felt necessary, e.g. due to practical problems and improved insights.

Those procedures, carried out by FIMCI to check the quality of data and information, can be divided in the following main steps:

- Registration and Documentation
- Inventory and validation checks
- Feedback Inventory Phase
- Digital storage of data and information

Information on these main steps has been given in the previous Technical Report (UN/ECE, EC; De Vries et al., 1998). Apart from checks on the consistency and validity of file names and file structures, the data integrity checks form the core of this data validation procedure. This includes on the validity of codes and the plausibility of results of parameters or parameter combinations. In view of the data submission with respect to soil solution chemistry, the data validation program FIMCI\_CK has been updated by including checks on the consistency of those data but also on deposition data. More specific information on those checks is given in the Sections 6.2.3 and 8.2.3.

#### *Data presentation*

In presenting the data, the temporal aggregation was set at one year for the continuous data with respect to deposition, meteorology and soil solution chemistry. For the soil solution chemistry parameters, a distinction was made between the organic layer and two mineral layers, namely a depth-weighted average up till 40 cm (biologically active topsoil that is most influenced by atmospheric deposition) and a depth-weighted average up between 40 cm and 80 cm, which is generally the lower boundary of the root zone (the zone in which at least 90% of the roots occur).

Regarding the assessment period, the year 1996 was used for all continuous monitoring data (crown condition, deposition, meteorology and soil solution). The data on increment, that are measured in 5 years intervals are limited to the most recent survey in the period 1990-1996. The number of data sets before 1994 is very limited. Data in the soil and foliar surveys are not presented since they have been extensively discussed in last year’s report.

## 3.2 Assessment of data reliability

### *General approach*

Within the Intensive Monitoring Programme a variety of data assessment methods is applied. Most important are variations in sampling layout and set-up (including sampling numbers), measuring equipment in the field and methods for digestion (soil and foliar survey) and analysis. As a result, differences in accuracy, reliability and representativity of the stored data exist. These aspects may influence the results of an evaluation and should be considered, e.g. by (i) a certain selection of the evaluation data set or (ii) giving higher weights to those data that are considered to be more reliable (see Section 3.3.2).

In last year's report (De Vries et al., 1998), the comparability of applied methods (as described in the DAR-Q's) has been focussed upon (i) the number of observations or samples that were taken (and possibly pooled in certain surveys) in a forest stand to get a representative value for that stand and (ii) the digestion and analyses methods that were used to measure the chemistry of foliage, soil and deposition. Here we only discuss the interpretation of the number of observations or samples at plot level in view of data reliability, which is a relevant aspect for all surveys. More specific information on the data assessment methods is given in the chapters on the respective surveys. This refers specifically to meteorological data (Chapter 7) and soil solution chemistry data (Chapter 8). Information with respect to the other surveys (crown condition, forest growth and atmospheric deposition) is limited to changes compared to the assessment in 1995, which has been described in last year's report.

### *Sampling numbers at plot level*

By examining the number of observations within the plot, used to assess the parameters in the various surveys (in combination with the sampling device that influences the size of the sample and the spatial variability in a stand), information was gained on the representativity of the measured values for the Intensive Monitoring plots. The number of observations (or samples) that are needed, in order to obtain a representative value of a certain parameter for a forest stand, depends mainly upon the spatial variability in the parameter concerned (relative standard deviation) and the required reliability (accepted margin of error) of the average value for a stand. In formula (Hammond and Mc Cullagh, 1978):

$$n = t_{\alpha}^2 \cdot S^2 / D^2 \quad (3.1)$$

where:

- n = number of observations (The observations equal the number of individual trees for each tree species cluster in the crown-, foliar- and increment surveys, the number of soil samples in the soil survey and the number of samplers in the deposition survey)
- $t_{\alpha}$  = tabled Student t factor for a given uncertainty  $\alpha$ ; for  $\alpha = 0.05$ ,  $t_{\alpha} = 1.96$
- S = standard deviation within the plot, relative to the mean value (%)
- D = margin of error within the plot, relative to the mean value (%)

Inversely, the margin of error, D, depends on the spatial variability, S, and the sample number according to:

$$D = t_{\alpha} \cdot S / \sqrt{n} \quad (3.2)$$

Depending on the required reliability, D, and the relative standard deviation in a given parameter, the required number of observations varies as given in Table 3.1 (see Eq. 3.1 with  $t_{\alpha} = 2$ ).

*Table 3.1 The required number of samples or observations at plot level for a given parameter as a function of the spatial variability of that parameter (relative standard deviation, S) and the required reliability (acceptable relative margin of error, D)*

Relative margin of error	Required number of samples				
	S=20%	S=30%	S=40%	S=50%	S=60%
D=5%	64	144	256	400	576
D=10%	16	36	64	100	144
D=15%	7	16	28	44	64
D=20%	4	9	16	25	36
D=25%	3	6	10	16	23

Table 3.1 shows that the number of samples/observations that is required to obtain an average value with a reliability within 5% is generally very high (generally >100), especially when the spatial variability is large. Such numbers are often considered impracticable. Accepting a margin of error of 20%, Table 3.1 indicates that a number of 5-36 samples is needed depending on the S-value. Using a number of 20-25 samples leads to a reliability of the average value within 10-25%, when the relative standard deviation varies between 20 and 60%. A relative standard deviation between 20 and 60% is a range that is often encountered for parameters in natural environments. For example, the soil solution chemistry of Dutch forest soils varied between the limits range depending upon the element considered. A similar variation, and related relative standard deviation, can be expected in crown condition parameters, deposition chemistry and meteorological parameters. The variation in forest growth data (tree height and diameter at breast height) is likely to be smaller, as they are less influenced by meteorological conditions (see De Vries et al., 1998). Consequently, the number of samples that is needed to obtain a representative value is probably lower. The actual number used are presented in the relevant chapters, where they are evaluated in view of the general approach.

In the evaluation of data in multiple regression analyses, the square root of the number of samples has been included as a weighting factor, to account for the reliability of the data (see Section 3.3.2 for further details).

### 3.3 Statistical data evaluation

#### 3.3.1 General approach

A statistical evaluation of the data was focused on the key parameters in the surveys on atmospheric deposition and soil solution chemistry. The studies were presently focused on the major ions in the deposition ( $\text{NH}_4$ ,  $\text{NO}_3$ ,  $\text{SO}_4$ , H, Ca, Mg and K) and in the soil solution (same ions including Al). These key parameters are strongly influenced by stand and site characteristics and meteorological data. Soil solution chemistry is furthermore influenced by soil chemistry. To test hypotheses about the impact of the various characteristics on atmospheric deposition and soil solution chemistry, statistical techniques were used to investigate the relationship between:

- Atmospheric deposition versus stand and site characteristics and precipitation.

- Soil solution chemistry versus stand and site characteristics, precipitation, atmospheric deposition and soil chemistry

Site characteristics included as predictor variables were soil type, geographic, ‘climatic’ or ‘deposition’ region and altitude. Stand characteristics included tree species, stand height, stand structure index and stand density index (See Section 5.2.2). More literature-based information on the predictor variables used is given in Section 6.2.4 and 8.2.4, respectively. Fig. 3.1 gives an overview of the expected relationships.

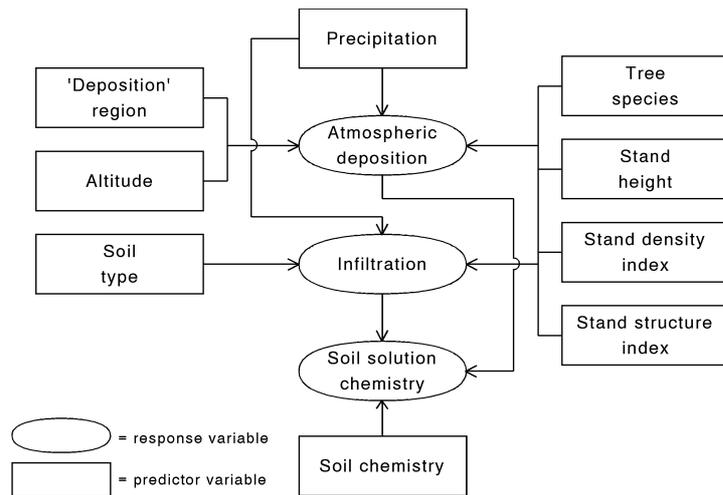


Figure 3.1 Expected relationships between the atmospheric deposition and soil solution chemistry versus stand and site characteristics, meteorological parameters and soil chemistry.

First insight in the possible correlations occurring at the investigated Intensive Monitoring plots can be obtained by applying ordination techniques. Such techniques give first insight in the correlations between all response variables (here the atmospheric inputs and the soil solution chemistry) and the predictor variables (here the stand and site characteristics, meteorological parameters and soil chemistry) as described below. More specific insight can be obtained by multiple regression techniques, relating the expectation value of a certain response variable to predictor variables in a quantified way. Both methods are described in more detail below.

### 3.3.2 Ordination techniques

Ordination is the collective term for multivariate techniques arranging sites on the basis of observed similarity in a variety of measured attributes (both response and predictor variables). It is also referred to as multidimensional scaling, component analysis, factor analysis and latent structure analysis (Ter braak, 1995). It can be used as an exploratory technique to analyse dose-response (cause-effect) relationships, being particularly suitable when analysing multiple resources (y-variables) to multiple causes (x-variables).

A distinction can be made in:

- Indirect gradient analyses: relates groups of sites with observed similarity in y (response) variables to associated x (dose or predictor) variables in an ordination diagram to (i) detect clusters of particular y-x combinations (similar sites) and (ii) determine the contribution of

each y- or x-variable to the imaginary variables (principal components) summarising those y- and x-variables (using  $R^2_{adj}$ ).

- Direct gradient analyses: relates the y-variables to linear combinations of x-variables, thus being comparable to (a reciprocal way of) multiple regression models (See Section 3.3.3).

The various multivariate techniques can further be distinguished on the basis of an assumed linear or optimal response between causes and effects. The latter approach is, for example, relevant when correlating environmental gradients in nutrients, water, acidity and temperature to species abundance. An overview of the various techniques, depending on the type of ordination and the assumed responses between the y- and x-variables is given in Table 3.2.

Table 3.2 *Multivariate techniques used in dependence of the type of ordination and the assumed responses between the y- and x-variables*

Ordinate technique (Gradient analysis)	Assumed linear response	Assumed optimal response
Indirect	Principal Component Analyses (PCA)	Correspondence Analysis (CA)
Direct	Redundancy Analyses (RA) Project to Latent Structure Analyses (PLS) <sup>1)</sup>	Canonical Correspondence Analysis (CCA)

<sup>1)</sup> Also denoted as Partial Least Squares Modelling

Application of multivariate techniques is particularly relevant when relating multiple responses, such as the chemistry of atmospheric deposition and soil solution, to multiple causes such as the stand and site factors influencing inputs and fates of pollutants.

In this context, principal component analysis (PCA) was used to determine meaningful patterns among the variables considered. Details of this method are provided by Geladi and Kowalski (1986). Geometrically the data points can be represented as points in a multidimensional space with the variables as axes. Distances and clusterings of points can be interpreted as similarities and dissimilarities among the variables. PCA calculates vectors (principal components) which fits best through the multidimensional data points. The first principal component is the vector of best fit for the data points. Subsequently, principal components can be calculated orthogonal to each other creating a plane or hyperplane and retain increasingly smaller  $R^2$ . To get an overview of the data set, two principal components are often sufficient. Subsequently identified principal components are characterised by a decreasing correlation coefficient, which usually becomes insignificant at the level of the third, or higher level component. The number of significant components is determined via cross-validation criteria given within the programme. An ordination diagram defined by 2 principal components (also called a biplot), can give relationships among the variables.

The PCA diagram (e.g. Figure 6.25) summarises the mutual correlations among response variables and, when measured, their correlation with predictor variables. In the PCA diagram, highly correlated variables lie close together, while uncorrelated variables lie far apart. Predictor variables can be superimposed on this plot, i.e. they are simply regressed on the sites after the construction of the diagram. Continuous predictor variables are represented by arrows, nominal ones by points. When a predictor variable point to a certain response variable their correlation is positive, when they point away from this variable, their correlation is negative. When their direction is perpendicular on the direction of the response variable they are uncorrelated.

### 3.3.3 Multiple regression techniques

Unlike ordination, regression analysis cannot be used to relate multiple responses to multiple predictors. Instead it focuses on a particular response variable and how this is related to (environmental) predictor variables. The term response variable stems from the idea that it responds to the environmental variables in a causal way, but causality cannot be inferred from regression analysis. The x-variables can either be selected from the indirect gradient analyses, being an exploratory method to gain insight in possible cause-effect relationships, or from hypothetically adopted cause-effect relationships based on literature information.

As a follow up of ordination, by ‘indirect gradient analyses’ (such as Principal Component Analysis, PCA), regression analysis is specifically suitable to:

- assess the relative contribution of environmental variables to the response variable, through tests of statistical significance.
- predict the response variable at sites where the environmental predictor variables are available.

Supposed relationships were of the form:

$$\log y = \alpha_0 + \alpha_1 x_1 + \alpha_2 x_2 + \dots + \alpha_n x_n \quad (3.4)$$

where  $\log y$  is the expectation value of the response variable (atmospheric deposition, soil solution chemistry),  $x_1$  to  $x_n$  are predictor variables (stand and site characteristics, meteorological parameters etc.) and  $\alpha_1$  to  $\alpha_n$  are the regression coefficients.

Some of the predictor variables were qualitative (indicator) variables, such as tree species and/or soil type. Interactions between two variables, implying that the effect of variable A depends on the level of variable B, were not included.

In order to meet the requirement of regression analyses that the response variable is normally distributed with a constant variance at fixed values of the predictor values, the considered responses were log-transformed. This also causes interaction to be less significant. Normality was checked by a scatter plot of the residuals against the fitted values.

The regression analyses was applied by using a so-called RSelect procedure. This approach combines forward selection, starting with a model including one predictor variable, and backward elimination, starting with a model including all predictor variables. The ‘best’ model was based on a combination of the percentage of variance accounted for ( $R^2_{adj}$ ), that should be high and the number of predictor variables, that should be low. Even though some site variables were intercorrelated, specifically (climatic) region and soil type, both were included since both do have a specific impact on most of the key parameters.

Further relevant aspects of the regression analyses are:

- Tree species, soil type and deposition region were included as qualitative variables. Based on the grouping described in the Technical Report of 1998 (De Vries et al., 1998) only 3 major groups of tree species were used (pine, spruce and broadleaves) and 3 main groups for soil type (Podzols and Arenosols, being acidic sandy soils, Cambisols and Luvisols being slightly acidic sandy soils and clayey soils and remaining non-calcareous soils). The limitation of tree species and soil types to 3 groups only was based on the expected differences in major influences, considering the limited number of data. With respect to atmospheric deposition, a

distinction was made in Northern Europe (divided in Boreal and Boreal Temperate), Western Europe (Atlantic climate), Central Europe (Continental and Sub-Continental) and Southern Europe (Mediterranean climate). Altitude and stand age, which are given in intervals of 50 m and 20 yr, respectively, were included as quantitative variables by using the average value of each considered class (e.g. 325 m in the altitude class 300-350 m and 30 yr. in the age class 20-40 yr.).

- The numbers of individual observations that were used to assess plot-mean values of the considered (log-transformed) key parameters were used as weighting factors in the regression analyses. This was done to give a weight to the reliability of the plot-mean value. The weighting factor was set equal to the square root of the number of observation, which is related to the margin of error of the plot-mean value (see Eq. 2 in Section 3.2). The observations equalled the number of samplers (funnels or gutters) in the deposition survey and the number of lysimeters in the soil solution survey. Information on those numbers is given in Chapter 6 and 8.
- Even though it may be relevant to include interactions between tree species and the other stand and site characteristics (since the effect of those characteristics on atmospheric deposition and soil solution chemistry may differ for various tree species), this was not done to limit the degrees of freedom used by the predictor variables. As a rule of thumb, the number of observations should exceed 4 times the degrees of freedom (Oude Voshaar, 1994).

Table 3.3 gives an overview of the predictor variables with their maximum degrees of freedom.

*Table 3.3 Overview of predictor variables and interaction included in the various regression models with their maximum degrees of freedom.*

Predictor variable	Degrees of freedom	
	Deposition	Soil solution chemistry
Tree species	3	3
Soil type	-	3
(Deposition) region	5	-
Altitude	1	-
Dominant height	(1)	-
Stand density index	(1)	-
Stand structure index	(1)	-
Atmospheric deposition	-	1
Precipitation	1	1
Soil characteristics	-	1-2
	10-13	9-10



## **4 Crown condition**

### **4.1 Introduction**

Crown condition, in terms of defoliation and discoloration is assessed at all monitoring plots. Both defoliation and discoloration are considered as key parameters, partly because this type of information has also been assessed at the systematic 16 x 16 km<sup>2</sup> grid in Europe since 1984. At the Intensive Monitoring plots, there are also a large number of additional parameters that may be assessed, but until now, only 3 countries submitted such optional data.

With respect to crown condition, it is important to concentrate the evaluation on changes over the years, since absolute crown condition data are influenced by differences in data assessment methods between the countries (Section 4.2.1). An in-depth interpretation is furthermore hampered by lack of information on stand history, pests and diseases, meteorological stress and air quality at most of the plots. The evaluation of crown condition data thus focuses on changes in crown condition, while checking the comparability of data assessment methods between 1996 and 1995. The evaluation is limited to defoliation, which is the most important parameter. Elevated discoloration values (discoloration class  $\neq 0$ ) were observed at a limited number of plots (<10%) and differences between 1996 and 1995 were small.

### **4.2 Methodological aspects**

In total 21 countries submitted DAR-Q's for crown condition assessed in 1995, which were stored in the database. The information given in these documents applied to 680 plots. In 1996 the number of plots for which DAR-Q Information was received raised to 700 plots (see Table 2.2). None of the participating countries that already submitted methodological information for 1995 reported any changes in methodologies by means of DAR-Q updates. The most relevant information in these DAR-Q's, being the number of assessment trees, the assessment methods and the assessment periods, has been presented in the previous Technical Report (De Vries et al., 1998).

In this report we focus on data comparability in view of differences in assessment periods and assessed trees in the years 1996 and 1995. Evaluations were carried out for 694 plots where data and methods for both 1995 and 1996 were available. This means that plots for which the data for one of these years lacked were not included (see Fig. 4.1).

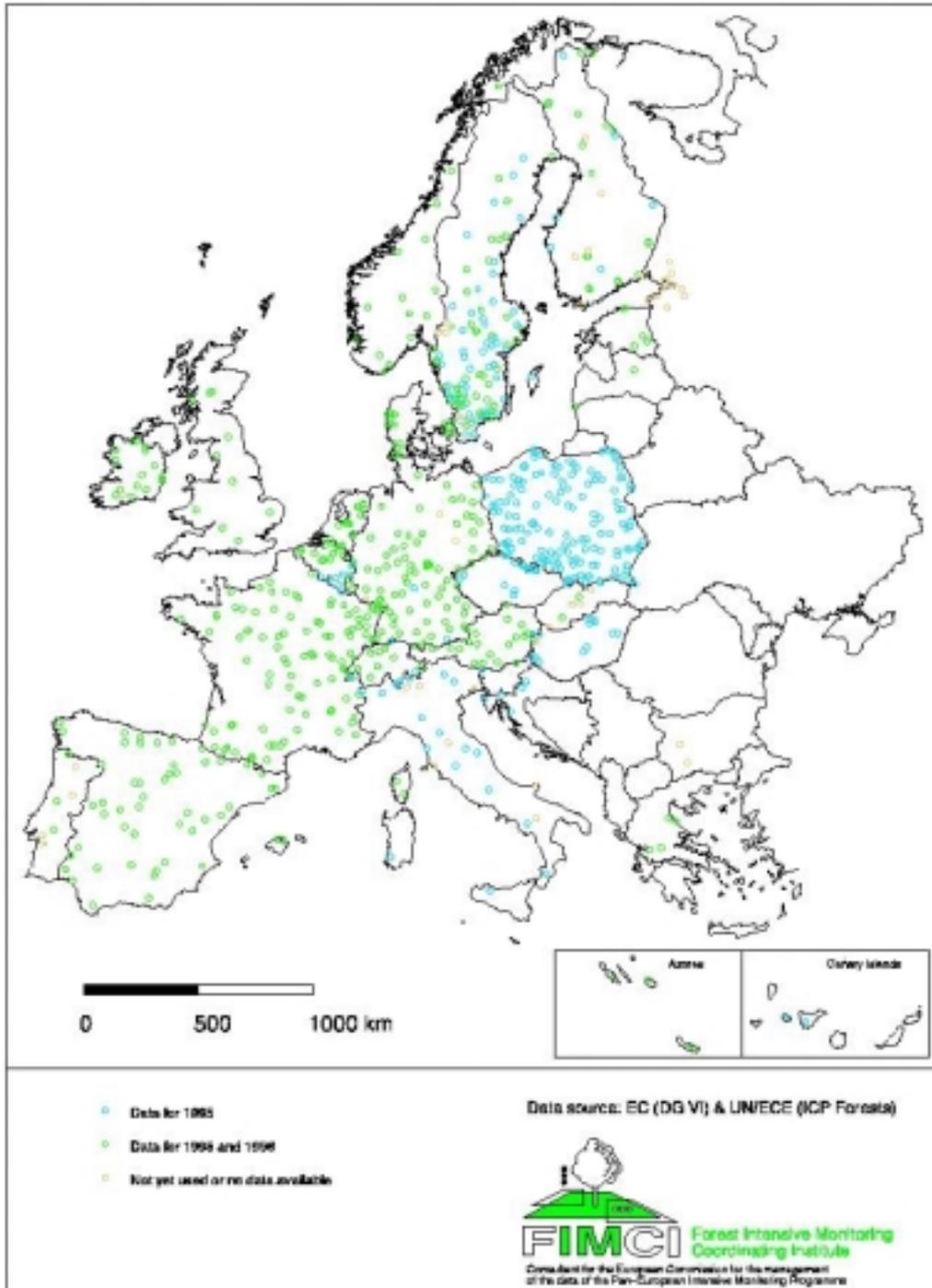


Figure 4.1 Geographical distribution of the plots of the Pan-European Intensive Monitoring Programme and years with crown condition assessments

Figure 4.1 Spatial distribution of the plots included in 1995 and 1996 crown condition assessments.

### **4.2.1 Locations**

Fig. 4.1 shows the spatial distribution of the plots at which the defoliation was assessed in either 1996 alone or in both 1995 and 1996. The map shows that at most plots the defoliation has been assessed in both years. A few plots were not assessed in 1996 (not separately depicted), although they were in the 1995 assessment. The map also shows that a considerable number of plots were only assessed in 1996. These plots are mainly located in Poland, the Czech Republic, Hungary, Croatia, Italy, Sweden and Wallonia. Most of these plots were assessed for the first time in 1996. Some of these plots, however, are assessed bi-annually, with 1994 being the previous assessment year.

### **4.2.2 Data assessment methods**

The assessment of crown condition depends on the definition of the reference tree and assessable crown and on the type of trees selected for the assessment. As stated before, the DAR-Q information for 1995 also applies to 1996 (no change in the assessment methods). In both years, on 75% of the plots, local reference trees were used, while on 25% of the plots absolute reference trees were used. Differences in definitions for assessable crown, that are likely to influence the values of assessed key parameters (defoliation, discoloration) and thus the data comparability, occurred similarly in both years (De Vries et al., 1998).

Finally, the selection of trees used to investigate crown condition was similar in 1995 and 1996. On 80% of the plots sample trees were in the Kraft classes 1-3 (pre-dominant, dominant and co-dominant). On 14% of the plots subdominant trees were also included in the sample population, whereas on 4% of the plots only predominant and dominant (Kraft classes 1 and 2) were assessed. On only 2% of the plots all Kraft classes were included.

### **4.2.3 Data comparability and data reliability**

#### ***Differences in the assessment periods***

Periods of assessment depended on tree species and climate and on specific weather conditions in the assessment year. Time of the annual reassessment should be adjusted according to the phenophase and actual weather conditions in the year. Consequently, it is not necessary that the assessment always occur exactly in the same period in different years (in this case 1995 and 1996). Results (Fig. 4.2) show that differences in assessment dates of less than one month occur at ca. 80% of the observations for broadleaves and ca. 70% of the observations for conifers (74% of all plots). About 14% of the all plots were assessed more than 30 days later, whereas 12% were assessed more than 30 days earlier. The largest changes were found for coniferous plots. In many cases these spectacular changes are due to adjustments in the prescribed periods in Manuals and Regulations. As such they imply an improvement in the data assessments. However, these changes may influence the comparability between 1995 and 1996 crown condition assessments.

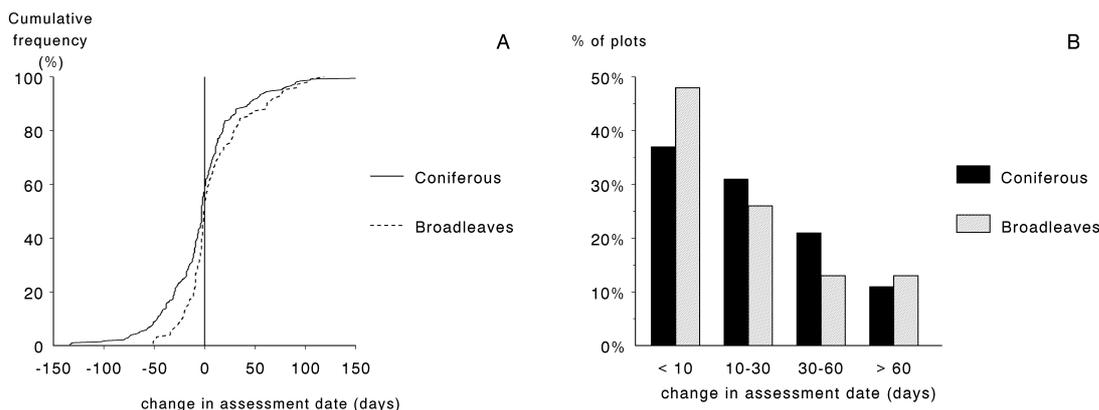


Figure 4.2 Differences in assessment periods for determination of defoliation and discoloration in 1996 and 1995 for broadleaves and conifers, presented as the cumulative frequency distribution of the differences (A) and the clustered numbers of the differences (B; earlier and later assessments combined).

For broadleaves, where the assessment period is of considerable importance for defoliation and discoloration assessment, more than 90% of the assessments were taken in July and August. The assessment periods ranged from end of June to the middle of October (Fig. 4.3). According to the received information the assessment for conifers ranged from February to December but most assessments occurred in July and August (Fig. 4.3). It is not certain whether such large variations in the assessment date affect the results, even if a deviating period is still within the period between the formation of new leaves/needles and the autumnal discoloration of leaves (like it has been formulated in Annex III of the Regulation). An assessment of conifers before the formation of new needles could even be considered as a late assessment date for an assessment in the previous year. Autumnal assessments (September or later) occurred relatively often for conifers. The autumnal assessments of broadleaves mostly refer to evergreen species, although a few assessments of deciduous species were even carried out in the beginning of October. Further investigation (e.g. by the EP on Crown Condition) of the possible impact of such deviating periods is recommended. For now, all results have been included in the evaluation.

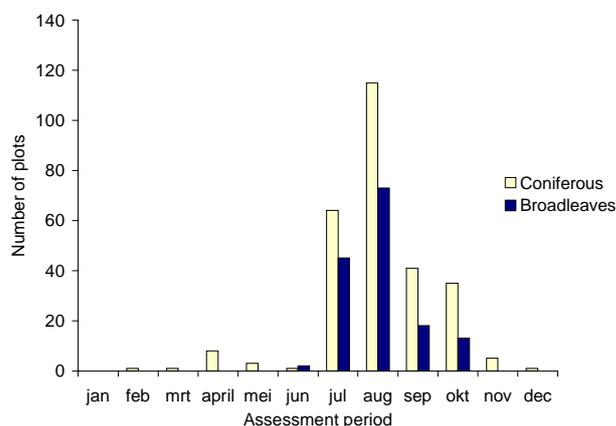


Figure 4.3 Assessment periods for determination of defoliation and discoloration in 1996 for broadleaves and conifers.

### Difference in the number of assessment trees

The range in the number of selected assessment trees in 1995 and 1996, which is of importance for the representativity of health status of the plot, is shown in Fig. 4.4. Results show that the same number of trees was used at more than 70% of the plots, with an additional proportion of

more than 10% with an increase or decrease of less than 10 trees. The number of stands with a considerable increase or decrease in number of assessed trees was very small.

An increase in the number of assessed trees was found at about 20 % of the plots, whereas a decrease was observed for less than 10% of the plots. This indicates that still efforts are made to improve the quantity and quality of the data from existing plots. The considerable increase in the number of assessed trees at some plots is probably related to changes in assessment strategy, which anticipate on the updates in the methodology described in the newest version of the manual.

In the 1996 crown condition assessments sample populations ranged from 15 up to 1056 assessment trees per plot. In 1995 the numbers of assessed trees ranged from 15 up to 1168. Only at 8 plots less than 20 assessment trees (given as minimum in the Manual for the assessment year 1996) were selected (Fig. 4.5). This number is comparable to 1995. On 48% of the Intensive Monitoring plots, selections of over 40 trees (indicated as ‘preferred’ in the Manual) were used. This group increased by 7% compared to 1995.

The number of assessed trees is likely to increase even further in future. Amendments in the EU Regulations and the ICP Forests Manual state that in principle all trees in the plot (or sub-plot) are to be assessed. These Amendments, however, do not yet apply to 1995 and 1996 crown condition assessments. As a result still many countries still measured a fixed number of (selected) assessment trees in these years.

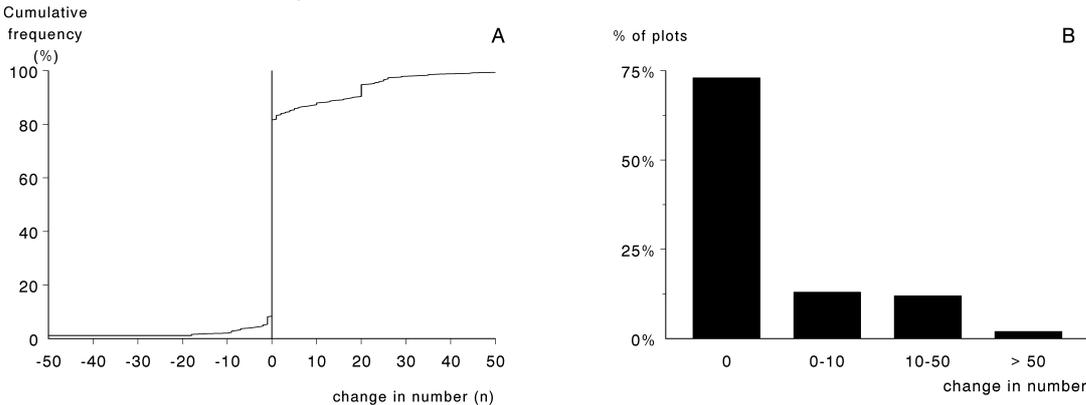


Figure 4.4 Comparison of the number of assessed trees per plot used in 1996 and 1995 for the determination of defoliation and discoloration presented as a cumulative frequency distribution (A) and a histogram (B).

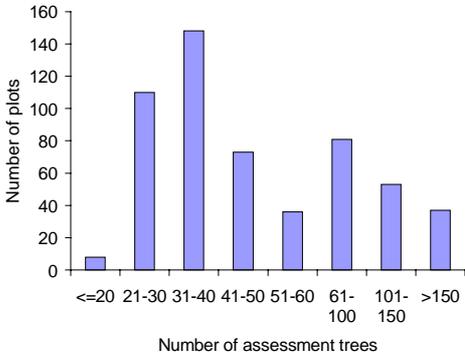


Figure 4.5 Number of assessed trees per plot used in 1996 for the determination of defoliation and discoloration

When only a few assessment trees are selected, it may have a bias effect as these trees may not represent the health status of the whole plot or forest ecosystem adequately, nor is the future situation (time series) sufficiently guaranteed (Eichhorn, pers. comm.). The number of trees that is required will depend on the within-plot variability in crown condition (Section 3.2). Information on this variability can be assessed for each plot since data are available at the tree level. Such information was given in the previous Technical Report, using the data for 1995 (UN/ECE, EC, de Vries et al., 1998). These results, which are comparable to 1996, showed a median value of 45% for the relative standard deviation and a median value of 21% for the margin of error. This shows that, on average, 20 sample trees is a minimum for a reliable plot-mean value of the defoliation (see Table 3.1). The reliability depends, however, on the (relative) standard deviation of the plot, which can be more than 100%. In this case several hundreds of trees are needed for a reliable assessment (see also Table 3.1). In general the variability in defoliation increases with age. For that reason it is relevant to assess crown condition for all trees in the plot as indicated in the amended EU regulation.

### ***Observer variability***

Another important issue related to the quality of crown condition data is the impact of stochastic observer variability and possible biases of observers. Information on such biases can be derived from joint field campaigns in which independent observers assess the crown condition at the same plots during the same period. No specific information for the Intensive Monitoring plots is available in this respect. However, results from investigations on reassessment sets from Austria, Finland, Norway, Switzerland and the United Kingdom (Mueller Edzards et al., 1997) learned that observer variation is an important factor when data quality is regarded. Whenever coarse classes (e.g. damaged vs. undamaged or the four damage classes) are used, the agreement between two individual assessments of defoliation was relatively high. However when small (5%) defoliation classes were used the agreement was relatively small (25-30%).

### **4.2.4 Data evaluation methods**

The presentation of results for 1996 and the differences between the 1996 and 1995 Crown Condition assessment was limited to the key parameter 'defoliation'. First, an overall evaluation for all plots was made using a grouping (clustering) of tree species in eleven groups, as described in De Vries et al (1998). This evaluation was based on the plot mean values per tree species, since the variation in the plot-mean values and the original values of the individual trees is very similar. The strongest deviation occurred in the range 0-10%, since trees with defoliation 0% were often mixed with more strongly defoliated trees, resulting in a lower proportion of values below 10% at the plot level (De Vries et al., 1998). The plot-mean values were calculated in a slightly different way than in previous years. The central value of each 5% defoliation class was used, instead of the class label (which is the upper limit of the class). For example, a given value 20% (= 16-20%) is now interpreted as 18%. The same method was also applied for the 1995 results, in order to make an appropriate comparison. The results of the plot-mean values in 1996 are first given as a function of the eleven tree species groups, using the distribution over the traditional defoliation classes (0-10%, 10-25%, 25-60%, 60-100%). The comparison with the 1995 results was limited to the plots for which data were available for both years.

A more in-depth comparison between the plot-mean defoliation values in 1996 and 1995 was carried out for the four major tree species (*Pinus sylvestris*, *Picea abies*, *Quercus robur/petraea*

and *Fagus sylvatica*) and for two typically Southern European species (*Quercus ilex* and *Pinus pinaster*). The four major tree species occur at most of the sites in the Intensive Monitoring Programme and have a broad geographic coverage. The two Southern European species are typical representants of the forest of Southern Europe, where the four major species are less common. The six selected species are the most important species from the following tree species clusters Pine (*Pinus sylvestris*), Spruce (*Picea abies*), Mediterranean Pine (*Pinus pinaster*), European Oak (*Quercus robur/petraea*), Beech (*Fagus sylvatica*) and Evergreen Oaks (*Quercus ilex*). In order to avoid confusion between the clusters and the individual tree species, the English names were used for the clusters, whereas the Latin names were used for the individual species.

## 4.3 Results and discussion

### *1996 results for all tree species groups*

The overall result of the 1996 defoliation assessment is illustrated in Table 4.1 by the proportion of stands in the various defoliation classes (using plot-mean defoliation values per tree species), distinguishing eleven major tree species groups. The number of plots included is considerable larger than for the 1995 assessment for almost all groups (compare Fig. 4.1).

The defoliation values for Conifers were generally lower than for Broadleaves (Table 4.1: ‘All Broadleaves’ vs. ‘All Conifers’). For Conifers the largest numbers of trees were found in the first two defoliation classes (0-10% and 10-25%), whereas most Broadleaved trees occurred in the 10-25% class, followed by the 25-60% class (Table 4.1). Only few stands were found in the classes with severe damage (60-100%) or dead. These high values were mostly related to plot-species-combinations with only a few individual trees, surrounded by trees of a different species.

Table 4.1 Distribution of the (plot-mean) defoliation values<sup>1)</sup> in 1996 at the Intensive Monitoring plots over the traditional defoliation classes

Tree species cluster	No of Plots <sup>2)</sup>	Proportion per defoliation class (%)				
		0-10%	10-25%	25-60%	60-100%	Dead
Pine	260	18.9	52.7	28.1	0.4	0.0
Spruce	231	31.6	44.6	22.9	0.4	0.4
Firs	51	31.4	31.4	33.3	3.9	0.0
Alpine Conifers	33	51.5	36.4	12.1	0.0	0.0
Mediterranean conifers	31	16.1	58.1	25.8	0.0	0.0
Remaining Conifers	8	37.5	25.0	37.5	0.0	0.0
Beech	121	28.9	38.0	27.3	4.1	1.7
European Oak	101	17.8	36.6	44.6	1.0	0.0
Evergreen Oaks	21	0.0	47.6	52.4	0.0	0.0
Other Oaks	24	8.3	54.2	37.5	0.0	0.0
Remaining Broadleaves	58	29.3	39.7	22.4	6.9	1.7
All Conifers	614	26.6	46.9	25.7	0.7	0.2
All Broadleaves	325	22.2	39.7	34.2	3.1	0.9
All	939 <sup>2)</sup>	25.0	44.4	28.7	1.5	0.4

<sup>1)</sup> Note that the results may be affected by the differences in assessment standards in the various countries.

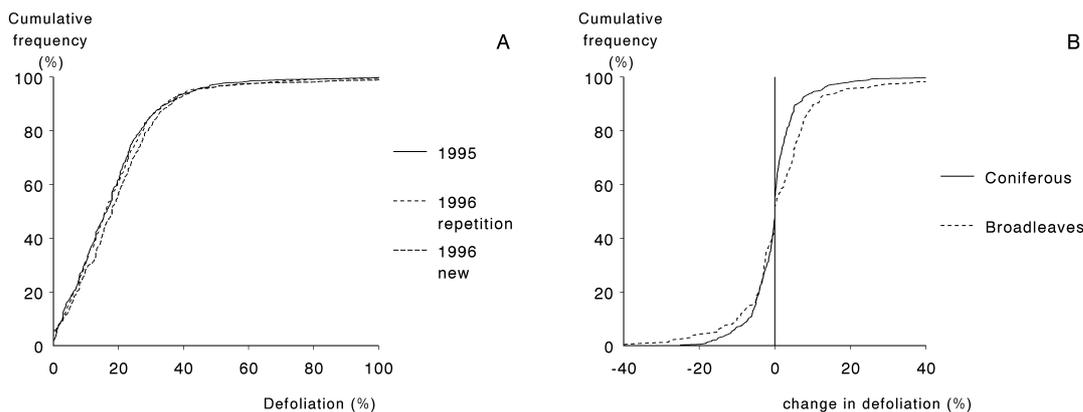
<sup>2)</sup> The number of observations is larger than the number of plots in ‘General Plot’, due to the occurrence of more than one species at several plots.

When considering the results as a function of the tree species clusters, the proportion of more or less healthy coniferous plots (average defoliation <25%) decreased from Alpine Conifers > Spruce & Mediterranean Conifers > Pine > Firs (Table 4.1). Within the Broadleaves the proportion of more or less healthy plots (average defoliation <25%) decreased from Beech > Other Oaks > European Oak > Evergreen Oaks. ). Like in the 1995 survey, the lowest proportions of

healthy trees (and thus the highest proportions of considerably defoliated trees) were found for the Mediterranean Conifers, the Evergreen Oaks and the Other Oaks. However, there seems to be an increase in the class slightly defoliated trees for Evergreen Oaks, which will be elaborated further-on, when the changes will be analysed pair-wise.

### ***Overall differences between 1995 and 1996***

The 1996 results for the 565 plot-species combinations that have been assessed in both 1995 and 1996 with the results of the 1996 assessment (Fig. 4.6A), whereas the overall differences between the 1995 survey and the 1996 survey also seem very small (compare Table 4.2; All Species). There are, however, considerable changes when subsequent observations at the same plot are analysed pair-wise. The plot-mean defoliation values generally changed less than 20% between 1995 and 1996 (Fig. 4.6). Larger changes are mostly related to plot-species combinations with one or only a few individual trees. The largest differences (both positive and negative) were found for broadleaves. Ca. 20% of the broadleaves and ca. 10% of the conifers showed an increase in defoliation of more than 10%, whereas ca. 15% of the broadleaves and ca. 10% of the conifers showed a decrease of more than 10%. These differences are further illustrated in Table 4.2, which gives an overview of the plot-mean defoliation values for the plots that have been assessed in both years.



*Figure 4.6 Overall comparison of the variation in the defoliation in the 1995 and 1996 assessment by cumulative frequency distributions of the plot-mean defoliation values (A) and the (pair-wise) changes between 1995 and 1996 for the coniferous and broadleaved tree species (B).*

Comparison of the 1995 and 1996 results, distinguished for the major tree species groups, shows, that there is a slight overall increase in defoliation (Table 4.2; All Conifers, All Broadleaves, All). There are, however, considerable differences between the various tree species groups. A considerable increase in the number of observations in the highest defoliation class was found for European Oak, and for a lesser extent also for Pine, Spruce and Fir. This means that, on average, there was a deterioration in crown condition for these species. Significant recovery (decrease in defoliation) was observed for the Alpine Conifers and Evergreen Oak, and for a lesser extent also for Mediterranean conifers and the two rest groups. More detailed results are given in Fig. 4.7 for the selected individual species.

Table 4.2 Changes in the distribution of the (plot-mean) defoliation values<sup>1)</sup> between 1995 and 1996 at the Intensive Monitoring plots (using the traditional defoliation classes) The values of the two worst classes have been combined with those in the class 25-60%, since only few values were found in these classes.

Tree species cluster	No of Plots <sup>2)</sup>	Proportion per class in 1995 (%)			Proportion per class in 1996 (%)		
		0-10%	10-25%	>25%	0-10%	10-25%	>25%
Pine	108	32.4	54.6	13.0	28.7	54.6	16.6
Spruce	148	33.8	47.3	18.9	31.8	48.6	19.6
Firs	46	32.6	37.0	30.4	32.6	34.8	32.6
Alpine Conifers	21	38.1	52.4	9.5	52.4	38.1	9.5
Mediterranean conifers	25	4.0	56.0	40.0	4.0	64.0	32.0
Remaining Conifers	8	37.5	12.5	50.0	37.5	25.0	37.5
Beech	80	18.8	42.5	38.8	27.5	37.5	35.1
European Oak	70	28.6	40.0	31.4	14.3	40.0	45.7
Evergreen Oaks	17	0.0	29.4	70.6	0.0	41.2	58.8
Other Oaks	13	0.0	53.8	46.2	0.0	53.8	46.2
Remaining Broadleaves	29	17.2	41.4	41.3	20.7	44.8	34.4
All Conifers	356	31.5	48.3	20.3	30.3	48.6	21.0
All Broadleaves	209	19.1	41.1	39.7	18.2	40.7	41.1
All	565 <sup>2)</sup>	26.9	45.7	27.5	25.8	45.7	28.4

<sup>1)</sup>Note that the results may be affected by the differences in assessment standards in the various countries

<sup>2)</sup>The number of observations is larger than the number of plots in 'General Plot', due to the occurrence of more than one species at several plots.

### Changes in defoliation for six major tree species

Changes in defoliation between 1995 and 1996, using the plot-mean values, are evaluated for the six considered major tree species (Fig 4.7) show that the changes for the coniferous species are generally smaller than for the broadleaved species. The stands of *Picea abies*, generally, showed an increase in defoliation (ca. 75% of the plots) between 0 and 10%, whereas for *Pinus sylvestris* and *Pinus pinaster* the number of increases almost equalled the number of decreases. Relatively large decreases (-20%) were found for *Pinus pinaster*. About 70% of the *Quercus robur/petraea* stand showed an increase in defoliation, with values up to +20%. The changes for *Fagus sylvatica* were generally small, but at ca. 10% of the plots a decrease in defoliation was found of more than 10%. In relative terms, more plots with a large decrease in defoliation were found for *Quercus ilex*. Here it seems that a considerable improvement of the health status of this species occurred between 1995 and 1996, although also a deterioration was observed at ca. 30% of the plots.

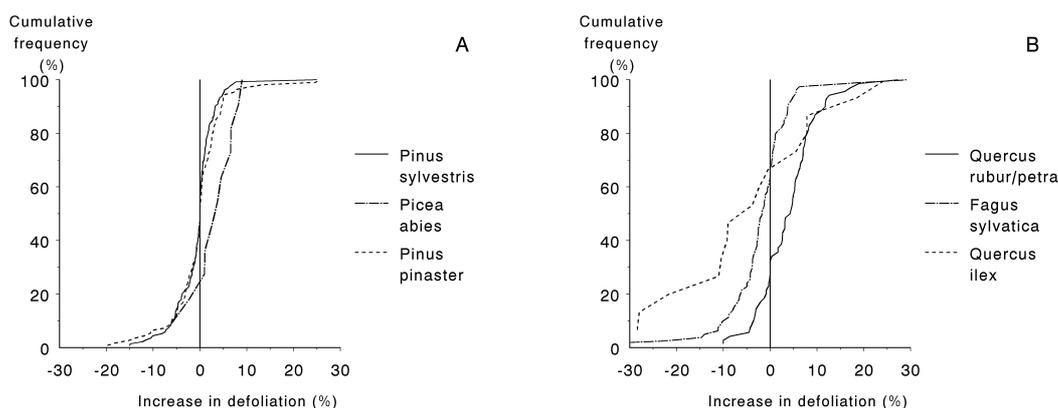


Figure 4.7 Changes in defoliation between 1995 and 1996 at the Intensive Monitoring plots covered with *Pinus sylvestris*, *Picea abies* and *Pinus pinaster* (A), and with *Quercus robur/petraea*, *Fagus sylvatica* and *Quercus ilex* (B). Positive values imply a decline and negative values an improvement of crown condition

#### **4.4 Conclusions**

The following conclusions can be drawn from the results discussed in the preceding sections:

- Data assessment methods were comparable between 1995 and 1996. The assessment dates in 1995 and 1996 differed less than one month for approximately 75% of the plots and the number of assessed trees remained the same at 72% of the plots. Most plots of the remaining 28% showed an (sometimes considerable) increase in the number of assessed trees, probably in anticipation on the amendments in the Regulation and Manual.
- On average, there was only a slight increase in defoliation between 1995 and 1996. However, a more or less significant deterioration in crown condition was observed for European Oak, Pine, Spruce and Fir, whereas a significant recovery was observed for Alpine Conifers, Mediterranean Conifers and Evergreen Oak.

The results do differ from those obtained at a systematic 16x16 km grid. This implies that the defoliation changes should not be used to present European wide overviews. Instead, those data are mainly useful in combination with other environmental (stress) factors to derive the relationships between them. Such a (preliminary) assessment is foreseen in next year's report.

## **5 Forest growth**

### **5.1 Introduction**

The (first) assessment of stand characteristics related to forest growth is driven by two main objectives:

- To provide baseline data for the analysis of variations in forest growth as a results of natural and anthropogenic growing conditions, including stand and site characteristics, soil chemical variables, meteorology and atmospheric deposition, when data of re-assessment become available.
- To provide data on stand characteristics, which can be used for the refinement of the relationship between these characteristics and different deposition and soil solution parameters.

One possible effect of elevated atmospheric deposition of N and S compounds is impact on forest growth. A positive effect can be expected in areas where forest growth is limited by N availability. Most likely, elevated N deposition increased forest growth in large parts of Europe (Spiecker et al., 1996). Inversely, a continuous high input of N leads to a situation where other growth factors, such as other nutrients and water, become limiting for the growth of forest. The relation between water shortage and N surplus can be explained by the fact that a high N input favours growth of canopy biomass, whereas root growth is relatively unaffected. The increase in canopy biomass will lead to a higher demand for water and therefore to an increased risk of water shortage (drought). It also causes an increased demand of base cation nutrients (Ca, Mg, K) whereas the availability of these cations can be reduced by increased dissolved levels of  $\text{NH}_4$  and/or Al (induced by  $\text{NO}_3$  and  $\text{SO}_4$ ).

Information on periodic annual increment at the Intensive Monitoring plots will only be available after the first re-measurement of the trees, five years after installation. Nevertheless, the data on tree diameter (at breast height) and tree height can already now be used to calculate a number of characteristics or indices related to site quality, stand density, stand structure and (tree) species diversity. Such stand characteristics do not only influence a considerable part of increment variation, but they also influence the input of atmospheric pollutants by differences in the scavenging of gases and aerosols (dry deposition). Furthermore, they may influence evapotranspiration and thereby soil solution chemistry (Section 3.3.3).

This chapter presents information on stand characteristics related to stand density, stand structure, (tree) species diversity and stand height by calculating them from data on tree number at the stand level and both diameter at breast height (dbh) and tree height at tree level, distinguishing between the tree species occurring at the plot.

### **5.2 Methodological aspects**

Information on data assessment methods has now been received for 440 plots. The overview of the results, however, is related to only 403 plots, for which also data were received. Comparison of information given in the DAR-Q's and in the data submission forms sometimes was not consistent. In these cases the information given in the data submission forms has been used.

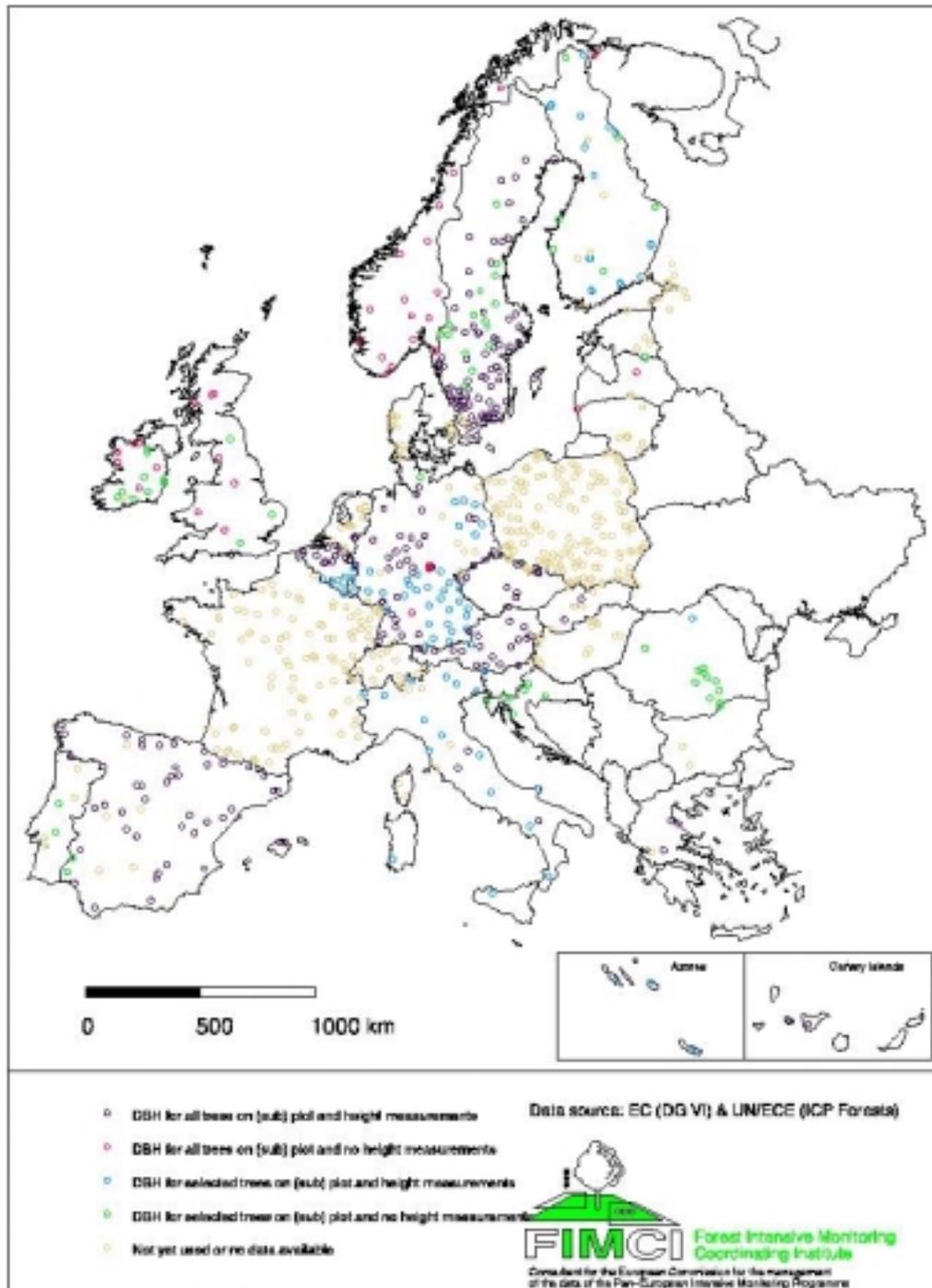


Figure 5.1 Geographical distribution of the plots of the Pan-European Intensive Monitoring Programme and parameters recorded in the increment surveys up to 1996

Figure 5.1 Spatial distribution of the plots included in the (most recent) forest growth assessments until 1996.

### 5.2.1 Locations

Fig. 5.1 gives an overview of the spatial distribution of the plots included in the Forest Growth inventory of the Intensive Monitoring Programme. All submitted and stored data until the 1996 survey have been included. Beware that some countries, such as France, submitted their first data with the 1997 data submission. These data are still in the validation process and are therefore not included in the present evaluation. The map shows in brown and red symbols the plots for which sufficient information was available (both from the data files and the DAR-Q) to calculate basal areas and stand density index (see Section 5.2.4).

Information on dbh measurements was also received for the plots depicted in blue and green, but this information was only measured at a selection of trees or it was not clear from the data files or the DARQ what the area was in which the dbh measurements were carried out. Apart from this, the map also gives an overview of the plots at which height measurements were carried out (brown and blue) or not (red and green). No restriction with respect to the number of height measurements was applied (cf. Section 5.2.4).

### 5.2.2 Data assessment methods

On all plots included in the forest growth survey, measurements of diameter at breast height (dbh) were carried out. On 32 % of the plots callipers have been used to measure the circumference of the trees, on 57 % of the plots tape was used. For 11 % of the plots no information was received concerning the applied methodology. On most plots, 'all' trees were assessed (either on the entire plot or on a subplot). On 11 plots (4%) a fixed number of trees has been measured for dbh and not all trees within a defined area. For these plots it is impossible to calculate values per area, e.g. basal area, based on the submitted information. Therefore the information from these plots has not been included in the evaluations described in section 5.3. A similar strategy had to be applied for a limited number of plots for which only part of the trees (or no trees at all) were labelled with a unique tree number. Trees need such a fixed unique number in the database to facilitate the comparison between subsequent Forest Growth assessment. Tree height was only measured at part of the plots and mostly for part of the trees, using Blume-Leiss and Suunto hypsometers.

### 5.2.3 Data reliability

Data reliability is influenced by the number of assessment trees. Fig. 5.2 gives an overview of the numbers of assessment trees for diameter at breast height (dbh) and tree height combined with the number of plots. Data presented apply to the most recent forest growth measurements at each plot.

dbh measurements were most frequently carried out on 'all trees' in a defined subplot (28 %) or 'all trees' in the whole intensive monitoring plot (23%). On average 140 trees per plot were measured, ranging from 24 to 1135 trees (see also Fig. 5.2A). For a relatively large number of plots it could not be determined whether plots or subplots have been used, either because files and/or DAR-Q's were incomplete (31%) or because the statements in files and DAR-Q's were contradictory (14 % of the plots). In the latter case the statements in the data files have been used. In a few cases the results for basal area and stand density index (see Section 5.3 for the

definitions) were not used because the values indicated that it was likely that not all trees at the (sub)plots were included.

If it was stated that 'all trees' were assessed, it was assumed that the trees of all Kraft Classes were included. The DAR-Q, however, does not give any additional information on the definition of 'all trees' and the applied selection criteria. Nor does it give detailed information on the use of a certain minimum diameter, the possible exclusion of certain 'weed species' or shrubs from the assessment, or the (physical) removal of understorey species, 'weed species' or shrubs during the installation or maintenance of the plot. Differences in these aspects might affect the results of the assessments. In the evaluation of the submitted data, however, it was assumed that such differences did not have a strong effect on the results.

At 83% of the plots where dbh was assessed also tree height was determined. It has to be noted that even measured tree height can be prone to large errors (Cluzeau et al., 1998). Approximately half of the countries determined tree height for all trees in the plot, whereas the other half reported measurements on a selection of trees (ranging between 8 and 50 trees). The average number of trees per plot at which tree height was measured, was 55. A maximum of 472 trees per plot was assessed and minimal 3 trees were measured (see also Fig. 5.2B).

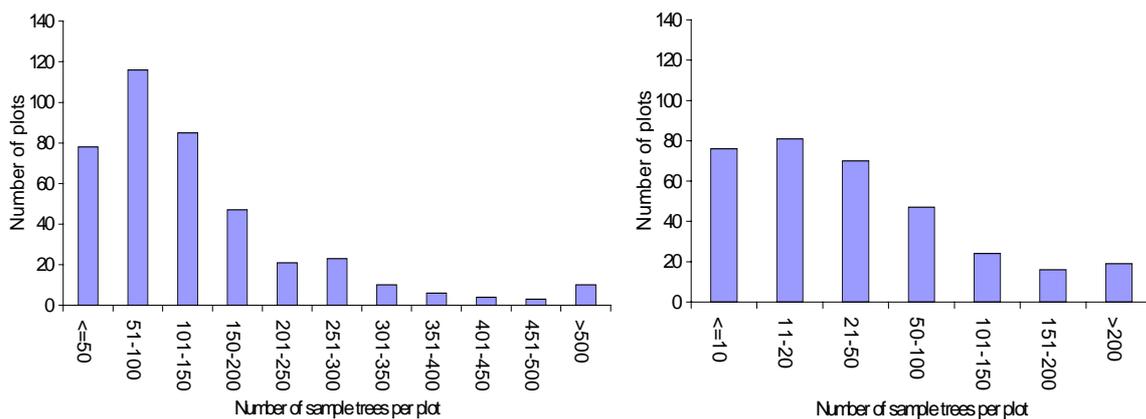


Figure 5.2 Number of sample trees used for the determination of diameter at breast height (A) and tree height (B).

As with crown condition, the number of trees that is required for a reliable assessment of dbh and tree height depends on the (spatial) variability. Calculations for each plot, based on the variation in results for individual trees, showed that the median values for the relative standard deviation,  $S$ , was 25% for dbh and 10% tree height. Due to the relatively large number of trees that were included in most assessments, the median margin of error,  $D$ , was 7% for dbh and 3% for tree height. Upper values were, however, as high as 109% and 77%, respectively. The comparatively large reliability of dbh and tree height is partly due to (i) the large proportion of even-aged stands (and monocultures) and (ii) the limitation to dominant trees, which is a relatively homogeneous subset with respect to these parameters.

#### 5.2.4 Data evaluation methods

Relevant stand characteristics influencing the forest growth, but also crown condition and/or atmospheric inputs include stand height, stand density and stand structure including species diversity. The calculation of relevant indices for these characteristics, based on dbh and height

measurements, is described below. The setup of this evaluation is partly based in a working document by Sterba (1998).

### ***Assessment of stand density***

Absolute measures of stand density include the tree number (N) and the basal area (BA) of the stand. The number of trees can vary considerable, related to the stand structure, the species composition and the stand treatment. The number of trees, as such, is therefore not a good measure to characterise the stand. The actual basal area also depends on various factors, such as tree species, stand age, site quality and stand treatment, but to a lesser extent than the number of trees. The basal area was calculated according to:

$$BA = \frac{\left( \sum_{i=1}^N dbh_i^2 \cdot \frac{\pi}{4} \right)}{PA} \quad (5.1)$$

where:

BA = basal area, being the sum of the area of the stems at breast height in a plot ( $m^2 \cdot ha^{-1}$ )

PA = plot area (ha)

dbh = diameter at breast height (cm)

Basal area was calculated for all separate tree species in each stand. These values were combined to a value for the basal area for the entire stand. The share of each species in the total basal area was used to gain insight in the dominance of trees in a stand. The species with the largest share was considered the dominant species. Stands were considered as monocultures when the dominant species contributed more than 80 % to the total basal area of the plot, and as a mixed stand if this contribution was less than 80 %. It was checked whether the given main tree species and the most common tree in the crown condition assessment also had the largest share in the basal area of the plot. Only for two plots the basal area of the given main tree species was not the largest at the plot.

The maximum basal area depends on stand age and site quality. In order to compare stands of different age and site quality, the information on the number of trees and the observed diameters was combined by calculating a stand density index according to (Reineke, 1933; Sterba 1987):

$$SDI = \frac{N}{PA} \cdot \left( \frac{25}{d_g} \right)^{-1.605} \quad (5.2)$$

with:

$$d_g = \left( \sum_{i=1}^N dbh_i^2 / N \right)^{0.5} \quad (5.3)$$

where:

$d_g$  = quadratic mean diameter (cm)

N = number of trees in the stand

The variable  $d_g$  is also referred to as diameter of the mean basal area stem, which follows from a combination of (Eq. 5.1) and (Eq. 5.3):

$$d_g = \left( \frac{BA}{N} \cdot \frac{4}{\pi} \cdot PA \right)^{0.5} \quad (5.4)$$

Reineke's stand density index thus normalises the tree number to a stand with trees with a dbh of 25 cm. The maximum of this index is supposed to be rather independent of stand age and site quality.

### ***Assessment of stand structure and species diversity***

As a measure of stand structure the coefficient of variation (cv) and the skewness (skew) of the dbh distribution was calculated for each plot, according to:

$$cv = \frac{1}{N} \cdot \frac{\left( \sum_{i=1}^N (dbh_i - \overline{dbh})^2 \right)^{0.5}}{\overline{dbh}} \quad (5.5)$$

$$skew = \frac{1}{N} \cdot \sum_{i=1}^N \left( \frac{dbh_i - \overline{dbh}}{sd} \right)^3 \quad (5.6)$$

where:

sd = standard deviation of the dbh distribution ( $sd = cv \cdot \overline{dbh}$ )

The more symmetric the distribution, the more uniform the stand appears. Such symmetric distributions are reflected in a skewness value close to 0, which is a typical characteristic of a even-aged monoculturous stand. Most uneven-aged stands (like Plenterwald) will have a positive skewness, related to a right-tailed distribution.

As a further measure of stand structure, the degree of tree species mixture was calculated according to the Simpson index (Simpson, 1949):

$$SI = 1 - \sum_{i=1}^S f_i^2 \quad (5.7)$$

where:

$f_i$  = the relative frequency of tree species i in the stand

S = the number of tree species in the stand.

Relative frequencies were based on (i) the tree number per species divided by the total tree number of the stand and (ii) the basal area per tree species divided by the total basal area of the stand. The latter value gives less importance to the admixture of species with a large number of thin stems, which are often considered less relevant for the forest structure. More information on the reliability of those data and the methods to derive stand characteristics from those data is given below.

### ***Assessment of mean and dominant stand height***

If the heights and diameters of all trees within a certain plot area are available, these can be used to calculate a diameter weighted (so-called Lorey's) mean stand height can, according to:

$$h_{Lorey} = \frac{\sum_{i=1}^N h_i \cdot dbh_i^2}{\sum_{i=1}^N dbh_i^2} \quad (5.8)$$

where:

N = number of trees in the stand (per species)

However, in most of the stands only part of the trees had been included in the tree height assessment. In such cases, a reasonable mean stand height can be estimate by using a height curve for each tree species based on the available combined data of dbh and height according to (Prodan, 1965)

$$h - 1.30 = \frac{dbh^2}{\alpha_0 + \alpha_1 \cdot dbh + \alpha_2 \cdot dbh^2} \quad (5.9)$$

where:

h = tree height (m)

This can be reformulated to the following expression, which was used in the fitting procedure.

$$Y = \alpha_0 + \alpha_1 \cdot dbh + \alpha_2 \cdot dbh^2 \quad (5.10)$$

with

$$Y = \frac{dbh^2}{h - 1.30} \quad (5.11)$$

At most of the plots (90% of a total of 361 plot-species combinations), those multiple regression analyses resulted in a percentage variance accounted for ( $R^2_{adj}$ ) above 86%, but mostly even above 95%. The estimated regression coefficients ( $\alpha_0$ ,  $\alpha_1$  and  $\alpha_2$ ) were then used to estimate the Lorey's mean height from the quadratic mean diameter:

$$h_{Lorey;estimated} = 1.30 + \frac{d_g^2}{\alpha_0 + \alpha_1 \cdot d_g + \alpha_2 \cdot d_g^2} \quad (5.12)$$

where:

$d_g$  = quadratic mean diameter (cm) (see Equation 5.3)

Similarly, Lorey's dominant height was calculated, applying Equation 5.8 to the 20% thickest trees in the stand, using dbh as the criterion. Dominant height is a good measure for site quality as

it is nearly independent of stand treatment (thinning). The calculation of Lorey's dominant height was restricted by a minimum number of height measurements at the 20% thickest trees. Even though Lorey's mean height and dominant height have also been calculated for the separate tree species occurring in mixed stands, results for the dominant height are only presented on a stand basis.

### ***Presentation of results***

The results for the various variables are presented first for the complete data set, by giving an overview of the variation in the plot total or plot mean values for the dominant tree species and for all trees together in the stand dominant tree species. Secondly, the results are presented as a function of common tree species groups (De Vries et al., 1998) by giving the mean values (and sometimes also the standard deviations) of the plot totals or plot means for these tree species. The same is done for the results for the dominant tree species only and for all separate secondary species. Finally, the variation between and within six major tree species is presented by cumulative frequency distribution of the plot total or plot mean values (for the entire stand) for these species. These six species include the four major tree species (*Pinus sylvestris*, *Picea abies*, *Quercus robur/petraea* and *Fagus sylvatica*) and two typically Southern European species (*Quercus ilex* and *Pinus pinaster*). The four major tree species occur at most of the sites in the Intensive Monitoring Programme and have a broad geographic coverage. The two Southern European species are typical representants of the forest of Southern Europe, where the four major species are less common.

Some additional results are presented, depending on the variable considered. The share of the Basal Area of the dominant, or main tree species is compared with the total Basal Area of the plot. The results on Basal Area, SDI and Skewness per tree species are compared with the dominance of the species at the plot. The Simpson Index (by Basal Area) is compared with the share of the dominant species in the Basal Area and with the Simpson Index (by numbers). Finally, the height of the dominant species is compared with its diameter.

## **5.3 Results and discussion**

### **5.3.1 Stand density**

#### ***Overall variation***

The stand density is characterised by the basal area (BA) and the stand density index (SDI). Both characteristics have been calculated per plot and per plot-species combination. The data for 253 plots could be taken into consideration (Table 5.1; compare Fig. 5.1).

The BA and SDI results show a considerable variation, both for the entire stand and when limited to the results of the dominant or main tree species (Fig. 5.3). The values at the lower end indicate that there are several relatively open stands, whereas the values at the upper end indicate that extremely dense stands do also occur. The dominant or main species mostly contributes at least 80 % to the total basal area of the stand (Fig. 5.4). About 50% of the stands completely consist of only one species (the vertical line at the left of Fig. 5.4). However, in approximately 20% of the stands, the dominant species contributes less, sometimes as low as 45%. The value of 80%

contribution has been used to separate stands that are almost monocultures from stands with a more or less mixed species composition.

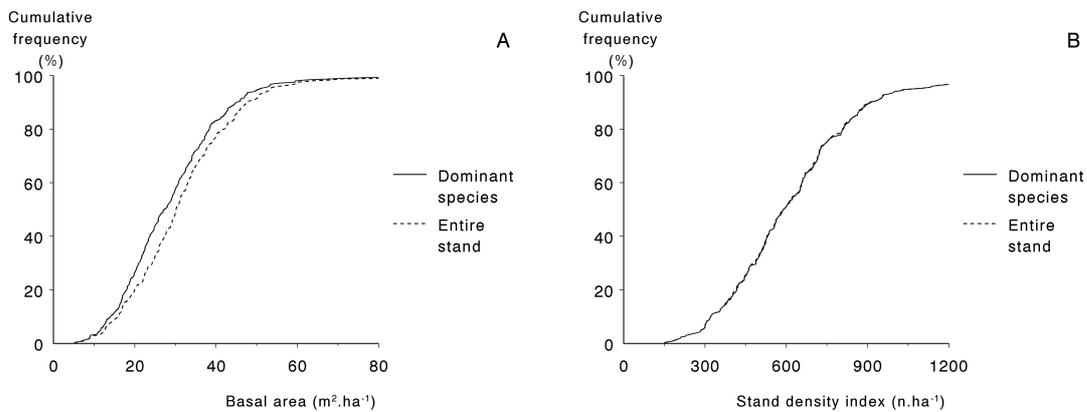


Figure 5.3 Cumulative frequency distributions of the Basal Area (A) and the Stand Density Index (B) at the Intensive Monitoring plots for entire stands and limited to the dominant/main tree species.

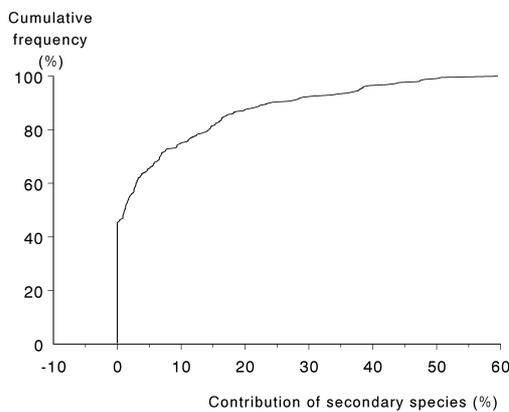


Figure 5.4 Cumulative frequency distribution of the contribution of secondary tree species to the entire Basal Area of the stand.

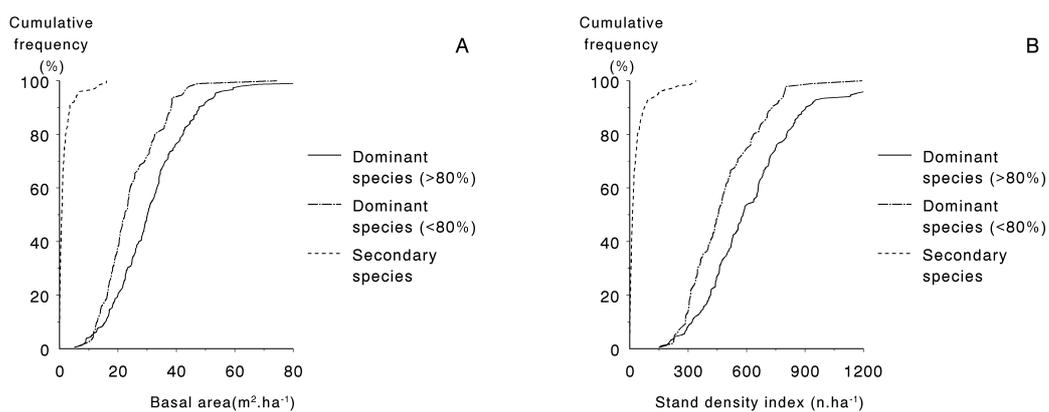


Figure 5.5 Cumulative frequency distributions of the Basal Area (A) and the Stand Density Index (B) as a function of the dominance of the species (see text).

Fig. 5.5 shows that the basal area and stand density index of the separate species is generally slightly higher for the dominant species in monocultures than for the dominant species in mixed

stands. The difference with secondary species is, however, much bigger. Those species generally contribute only marginally to the basal area and the stand density index of the entire stand.

### ***Variation between the species groups***

Pine and Spruce stands are most common most in the available data on basal area and stand density index with 45 and 110 stands, respectively (Table 5.1). Relatively large mean BA and SDI values were found in stands dominated by Spruce. Relatively low mean values were found for the three groups with Oaks. The high values for Fir and Remaining Broadleaves and the low values for Remaining Conifers, are possibly affected by the small number of observations.

*Table 5.1 Variation in the basal area (BA) and the stand density index (SDI) of entire stands at the Intensive Monitoring plots as a function of the main tree species*

Tree species cluster (by main tree species)	N	Basal area (m <sup>2</sup> ha <sup>-1</sup> )		Stand density index (n ha <sup>-1</sup> )	
		Avg.	St.Dev.	Avg.	St.Dev.
Pine	45	31.7	18.9	625	240
Spruce	110	34.9	14.6	696	268
Firs	1	42.9		716	
Alpine Conifers	4	27.9	8.1	625	150
Mediterranean conifers	19	28.8	11.9	541	206
Remaining Conifers	1	9.1		229	
Beech	32	31.8	7.2	563	145
European Oak	16	25.9	5.6	514	136
Evergreen Oaks	12	21.5	13.0	444	276
Other Oaks	9	21.0	9.3	485	227
Remaining Broadleaves	4	53.7	35.6	1153	694
All Conifers	180	33.2	15.5	658	258
All Broadleaves	73	28.7	13.2	555	271
All	253	31.9	15.0	628	265

Similar (but slightly lower) results were found when the results for the dominant species only were used (Table 5.2; left). Further investigation of the results for the secondary species showed, that Pine and Remaining broadleaves occurred at many plots as a secondary species. However, the BA and SDI for the secondary Remaining Broadleaves are generally low, which indicates that they do not contribute significantly to the stand structure and the forest biomass (Table 5.2; right). This cluster probably includes a number of small or shrub-like tree species. A significant contribution of secondary species in the BA and SDI was found for Pine, Spruce and Firs and to a lesser extent also for Alpine Conifers and Beech. The occurrence and contribution of secondary species is strongly related to the forest management system, which needs more attention when forest growth and the spontaneous and guided forest development will be studied in more detail.

### ***Variation within the major tree species***

The observed variation in the basal area and SDI is larger for the stands dominated by the coniferous major tree species compared to those dominated by broadleaved species (Fig. 5.6). Within the coniferous stands, the results for *Pinus sylvestris* and *Pinus pinaster* show a similar distribution. A relatively large contribution of densely stocked stands was observed for the stands dominated by *Picea abies*. This is in line with the differences in ecological behaviour of the considered tree species, with respect to the normal levels of biomass and light transmission of the canopy. Within the broadleaved stands, the stands dominated by *Fagus sylvatica* generally showed the largest values for both basal area and SDI, which is also in line with the expectations. The stands dominated by *Quercus robur* or *Quercus petraea* generally showed little variation in BA and SDI. Most stands dominated by *Quercus ilex* are relatively open, although 20% of these

stands were even more densely stocked than the stands dominated by *Fagus sylvatica*. This variation is in line the natural variation and with the different management systems applied for these forests.

Table 5.2 Variation in the basal area (BA) and the stand density index (SDI) of separate species at the Intensive Monitoring plots as a function of the main tree species and secondary tree species

Tree species cluster	Main/dominant species			Secondary species		
	N <sup>1)</sup>	BA (m <sup>2</sup> ha <sup>-1</sup> )	SDI (n ha <sup>-1</sup> )	N <sup>2)</sup>	BA (m <sup>2</sup> ha <sup>-1</sup> )	SDI (n ha <sup>-1</sup> )
Pine	45	29.9	575	51	4.0	75
Spruce	110	31.3	630	28	4.8	99
Firs	1	42.9	716	17	4.2	79
Alpine Conifers	4	23.5	529	17	3.7	61
Mediterranean conifers	19	28.6	537	1	1.1	18
Remaining Conifers	1	9.1	229	2	0.1	3
Beech	32	29.3	516	25	2.4	51
European Oak	16	21.8	417	22	1.9	32
Evergreen Oaks	12	21.4	442	2	1.3	31
Other Oaks	9	20.9	481	5	0.2	5
Remaining Broadleaves	4	48.2	1026	105	0.9	18
All Conifers	180	30.4	603	116	4.1	77
All Broadleaves	73	26.3	506	159	1.2	25
All	253	29.2	575	275	2.4	47

<sup>1)</sup> One species for every plot; so the numbers are equal to those in Table 5.1.

<sup>2)</sup> Based on zero to several secondary species per plot (separately).

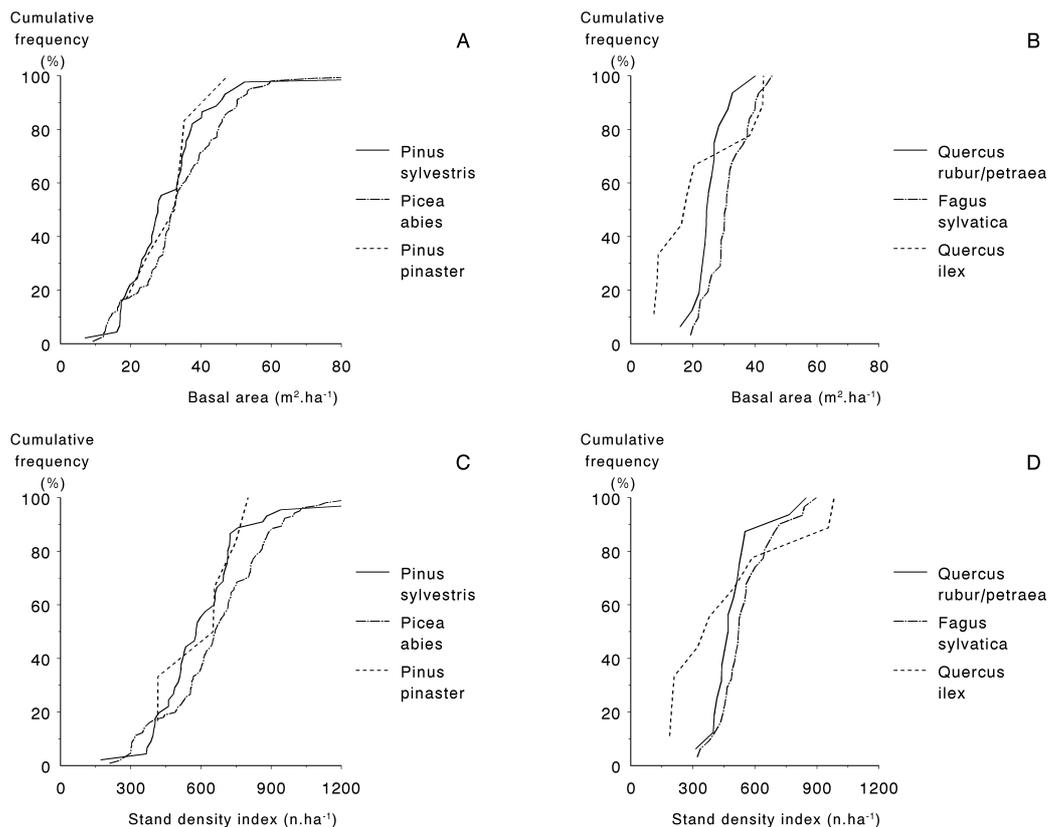


Figure 5.6 Basal Area (A&B) and Stand density Index (C&D) of the entire stand at the Intensive Monitoring plots dominated by *Pinus sylvestris*, *Picea abies* and *Pinus pinaster* (A&C), and by *Quercus robur/petraea*, *Fagus sylvatica* and *Quercus ilex* (B&D).

### 5.3.2 Stand structure and species diversity

The stand structure and species diversity are characterised by the skewness of the distribution of the measured diameters and by the Simpson index, respectively.

About 85% of the skewness values for the diameter distribution of entire stands and the dominant tree species occurred between  $-1$  and  $+1$  (Fig. 5.7A). This indicates that most of the observed stands have a uniform structure. Approximately 60% of the values was larger than 0, whereas 40% was lower. This indicates that there are more stands with positive outliers (relatively thick trees) than stands with negative outliers (relatively thin trees). This is a typical pattern for uniform, even-aged stands with many trees with a similar dbh and a few thick trees. Approximately 10% of the stands had a skewness larger than 1, which indicates uneven-aged stands with a decrease in number of stems with increase in dbh. This is substantiated by the results for the different age classes (Table 5.3), which shows the highest mean skewness for the uneven-aged stands. Only a few stands showed a skewness smaller than  $-1$ , which would be an indication for a stand with a second storey, which is less abundant than the trees in the dominant canopy. As expected, the youngest stands appeared to have the most uniform dbh distribution.

The results for the Simpson's Indices for species diversity show that almost 50% of the stands consist of one species exclusively (Fig 5.7B; Index = 0). The Index by basal area is generally slightly lower than the Index by stem numbers, which indicates that the species mixed with the main species contribute less to the basal area than to the number of stems. This means that the trees of the secondary species are generally considerably thinner than the trees of the main tree species. Only a few stands appeared to have a really mixed species composition: only ca. 5% of the stands had a Simpson's Index (by BA) above 0.50.

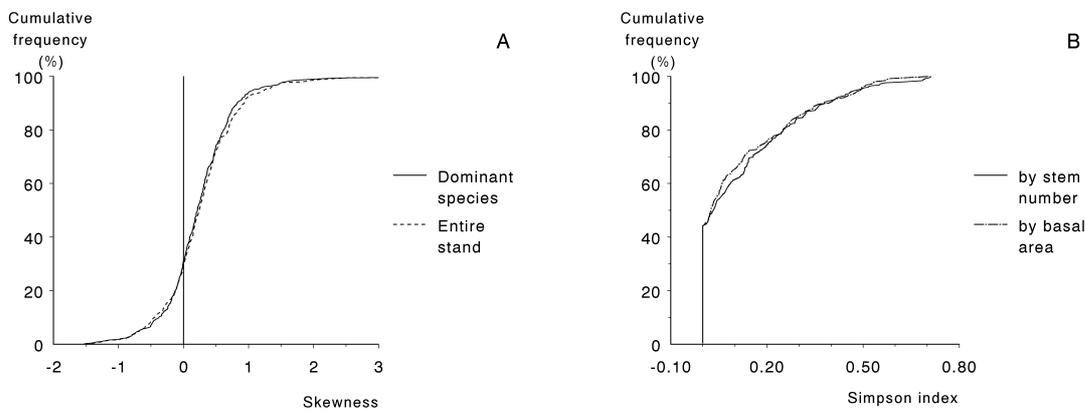


Figure 5.7 Cumulative frequency distributions of the skewness of the dbh distribution for entire stands and dominant species (A) and of the Simpson's Index of species diversity by stem number and basal area (B)

Table 5.3 Variation in skewness of the diameter distribution of the entire stand as a function of the mean age class.

Age Class	Number of stands	Mean skewness
0 – 20 years	7	0.06
20 – 40 years	40	0.41
40 – 60 years	102	0.24
60 – 80 years	99	0.21
80 – 100 years	51	0.30
100 – 120 years	42	0.20
> 120 years	38	0.27
Uneven aged	19	0.92

Further investigation of the skewness of the diameter distribution per plot-species combination by separating the trees in the earlier distinguished classes of dominance (see Section 5.3.1) shows that there was hardly any difference in the distribution of skewness values for the two classes of dominant species (Fig. 5.8). The skewness for the secondary species was generally larger, which indicates that most secondary species had a right-tailed diameter distribution with a decrease in abundance with increasing diameters. This might be an indication that a considerable part of the secondary or understorey species originate from natural regrowth.

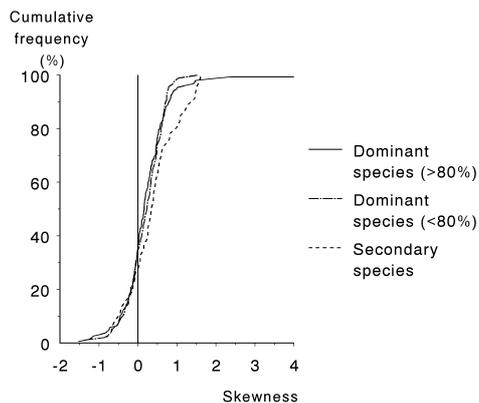


Figure 5.8 Cumulative frequency distributions of the skewness (per species) as a function of the dominance of the species.

### ***Variation between the species groups***

The values of the skewness for the various tree species groups shows that the stands dominated by Pine, Alpine Conifers, Mediterranean Conifers and Beech had a relatively homogeneous distribution of dbh values (Table 5.4; left; skewness  $< \pm 0.20$ ). The stands dominated by conifers were generally more homogeneous than the stands dominated by broadleaves. The homogeneity of the broadleaved stands, however, increased considerably when only the dominant tree species was taken into account. This is most noticeable for the stands dominated by European Oak. This means that many of these stands were mixed with species with deviating dbh values. This may also indicate that the stands dominated by European Oak generally had a considerable admixture of secondary species, which is substantiated by the results of the Simpson Index (Table 5.5).

Particularly high values for the Skewness were found for the stands dominated by Evergreen Oaks, and for a lesser extent for Other Oaks and Remaining Broadleaves (Table 5.4). This is probably related to the high proportion of uneven-aged stands for these tree species. The results for the secondary species show relatively high values (with enough observations) for Beech and Remaining Broadleaves, which indicate that these species contribute significantly to the structural diversity in the understorey of the stands in which they appeared as a secondary species. Relatively high values for the Simpson's species diversity indices ( $> 0.10$ ) were also found for Pine, Spruce, Alpine Conifers, Beech and Remaining Broadleaves (besides Evergreen Oak).

Table 5.4 Variation in the skewness in observed diameters of entire stands and separate species at the Intensive Monitoring plots as a function of the main or considered tree species.

Tree species cluster	Entire stand (by main spec.)			Separate species			
	N <sup>1)</sup>	Avg.	St.Dev.	Dom./main species		Secondary species	
				N <sup>2)</sup>	Avg.	N <sup>3)</sup>	Avg.
Pine	80	0.12	0.53	69	0.26	25	-0.10
Spruce	158	0.29	0.79	149	0.27	21	0.56
Firs	3	0.72	0.77	3	0.07	12	0.52
Alpine Conifers	6	-0.17	0.45	6	-0.06	9	-0.06
Mediterranean conifers	21	-0.12	0.57	21	-0.14	0	
Remaining Conifers	5	0.30	0.60	5	0.29	1	1.56
Beech	56	0.18	0.55	54	0.14	20	1.09
European Oak	31	0.46	0.54	27	0.12	9	-0.06
Evergreen Oaks	16	1.10	1.04	16	1.04	1	3.54
Other Oaks	15	0.66	0.62	12	0.65	6	0.53
Remaining Broadleaves	7	0.70	0.79	6	0.75	75	0.77
All Conifers	273	0.21	0.71	253	0.22	68	0.24
All Broadleaves	125	0.45	0.71	115	0.34	111	0.77
All	398 <sup>1)</sup>	0.28	0.72	368 <sup>2)</sup>	0.26	179 <sup>3)</sup>	0.57

<sup>1)</sup> One value for every plot with sufficient data (5 dbh measurements). No limitations were made with respect to the availability of information on the plot size etc. Therefore, the number of observations included is larger than for Basal area, Simpson Index etc.

<sup>2)</sup> One value for the dominant species for every plot with sufficient data (5 dbh measurements at the dominant species). The number of stands included may be smaller than for the entire stand in case the number of dbh measurements for the dominant species is less than 5, whereas the number of dbh measurements for the entire stand (irrespectively of species) is more than 5.

<sup>3)</sup> Based on zero to several secondary species per plot (separately) with sufficient data (5 dbh measurements).

Table 5.5 Variation in the Simpson's species diversity index (by number of stems and by contribution to the basal area) as a function of the main tree species.

Tree species cluster (by main tree species)	N (reps.)	Simpson index (by # of stems)		Simpson index (by basal area)	
		Avg.	St.Dev.	Avg.	St.Dev.
Pine	45	0.14	0.16	0.10	0.14
Spruce	110	0.12	0.16	0.14	0.17
Firs	1	0.00		0.00	
Alpine Conifers	4	0.13	0.20	0.15	0.24
Mediterranean conifers	19	0.02	0.08	0.01	0.02
Remaining Conifers	1	0.00		0.00	
Beech	32	0.11	0.14	0.11	0.15
European Oak	16	0.32	0.28	0.24	0.22
Evergreen Oaks	12	0.01	0.02	0.01	0.02
Other Oaks	9	0.04	0.09	0.01	0.03
Remaining Broadleaves	4	0.26	0.32	0.18	0.26
All Conifers	180	0.12	0.15	0.11	0.16
All Broadleaves	73	0.14	0.21	0.12	0.17
All	253	0.12	0.17	0.11	0.16

### Variation within the major tree species

Further investigation of the results of the skewness of the diameter distribution and of the dominant species of the six selected species and the Simpson Index for these same stands, shows that stands dominated by *Pinus pinaster* or *Quercus ilex* were hardly ever mixed with other species (Fig. 5.9C&D). These, however, are very different with respect to the skewness of the diameter distribution. All *Quercus ilex* stands had a positive skewness (Fig. 5.9B), which indicates that these stand mostly had a 'normal' or 'natural' right-tailed diameter distribution. On the contrary, the skewness of the *Pinus pinaster* stands was generally clearly negative (Fig. 5.9A), indicating the these stand generally consisted of a uniform set of thick stems with a distinct proportion of thinner individual (possibly underpressed or in the understory).

The distribution of the skewness results for *Fagus sylvatica* shows a similar, but less strong, pattern as *Pinus pinaster* (Fig. 5.9A), which also could be an indication that a considerable proportion of the stands of this species consist of a dominant layer of this species with a limited, but distinct number of trees of the same species in the understorey. The results of the skewness for the remaining species shows a similar distribution as the overall distribution (Fig. 5.9A&B; compare Fig. 5.7A).

Approximately 40 to 50% of the stands dominated by *Pinus sylvestris*, *Picea abies* and *Fagus sylvatica* had Simpson Index of 1, which means that these stands were pure monocultures. The remaining 50 to 60% values of the Simpson Index for these species are more or less equal distributed between 0 and 0.3. Only 20% of the stands dominated by *Quercus robur* or *Q. petraea* were pure monocultures (Table 5.9D). Moreover, approximately 40% the stands dominated by these species had a Simpson Index between 0.1 and 0.35, which indicates that these are the stands with the strongest character of a mixed forest, of all stands included in the programme. This reflects the result in Table 5.5.

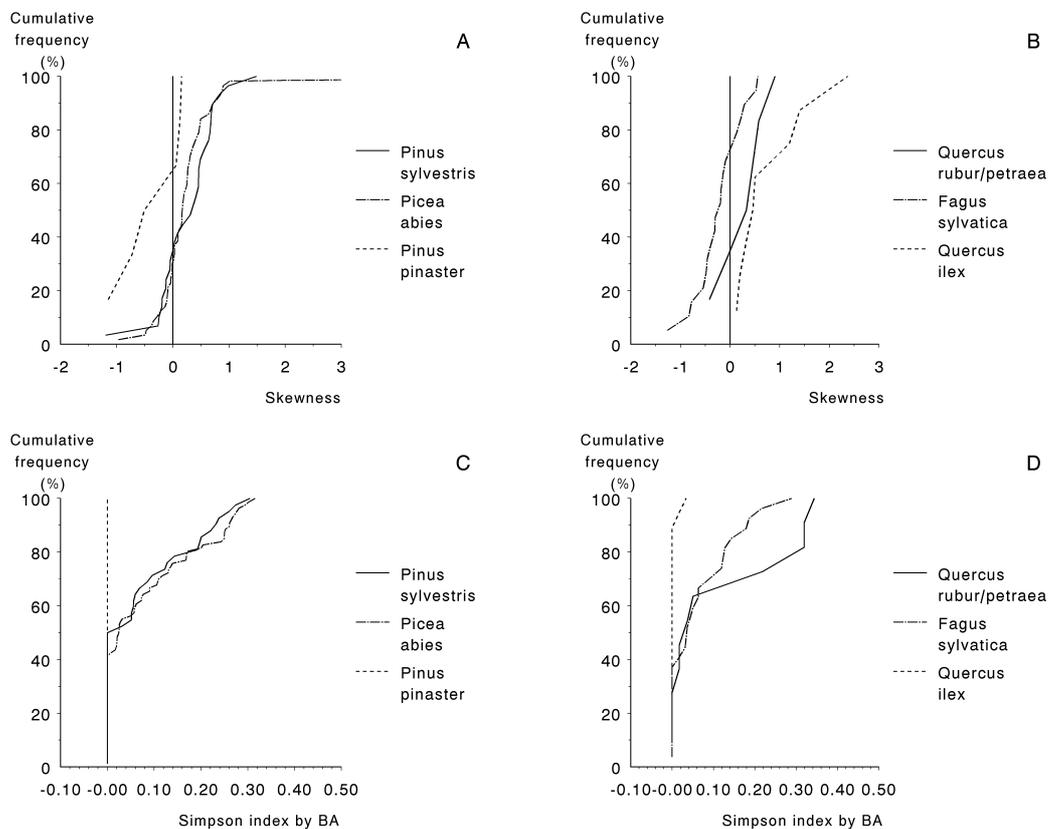


Figure 5.9 Cumulative frequency distribution of the skewness of the diameter distribution (A&B) and the Simpson's species diversity index by basal area (C&D) at the Intensive Monitoring plots dominated by *Pinus sylvestris*, *Picea abies* and *Pinus pinaster* (A&C), and with *Quercus robur/petraea*, *Fagus sylvatica* and *Quercus ilex* (B&D).

### 5.3.3 Stand height

#### Overall variation

Relevant characteristics of the stand height (Lorey's mean height (per species) and Lorey's dominant stand height) are estimated from the height measurements at a selection of trees in each stand. Fig. 5.10 shows the variation in the calculated values for both variables. Lorey's mean height could be calculated for the main species in 253 plots and for a limited number of secondary species, depending on the number of measurements. Lorey's dominant stand height could only be calculated for 140 stands, since sufficient height data had to be available for the subset of trees with the largest diameters.

Fig. 5.10A and B show that there was a considerable variation in both the Lorey's mean height and Lorey's dominant stand height. Both measures are strongly affected by the age of the stand and the site quality, whereas the mean height is also affected by the management system. Lorey's dominant stand height was nearly always larger than Lorey's mean height when comparing the data from the same stands (Fig. 5.10B), which is in line with the expectations that a subset of thick trees would also be taller. Furthermore, both variables showed an increase with increasing age (Table 5.6). Furthermore, the values of both variables increased with age, which is in line with the expectations that the trees grow and the height of the forest increases with age (Table 5.6).

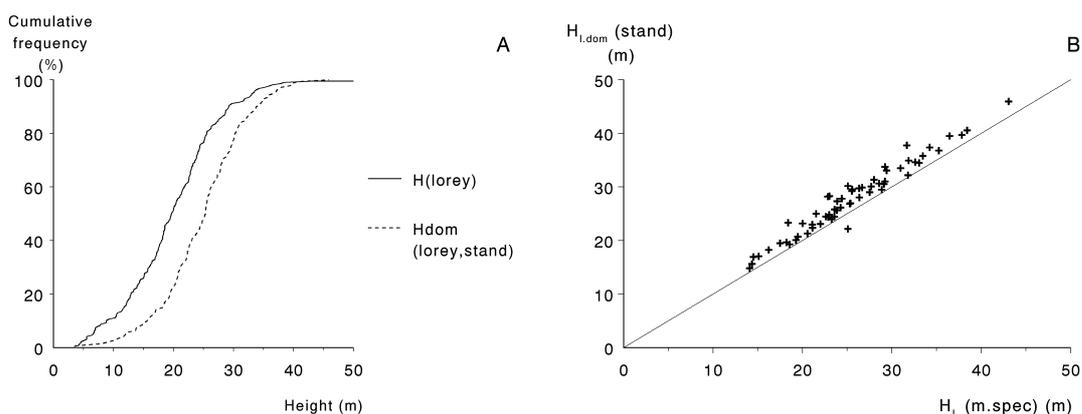


Figure 5.10 Variation in (A) and comparison of (B) the results for the Lorey's mean height of the main tree species and the Lorey's dominant height of the entire stand at the Intensive Monitoring Plots.

Table 5.6 Variation in the Lorey's mean height of the main tree species and the Lorey's dominant height of the entire stands as a function of the mean age class.

Age Class	H <sub>Lor</sub> of main species		H(dom) <sub>Lor</sub> of entire stand (by main spec.)	
	Number of stands	Mean values	Number of stands	Mean value
0 – 20 years	1	11.3	3	6.9
20 – 40 years	13	12.3	8	18.2
40 – 60 years	60	17.2	29	21.2
60 – 80 years	59	19.5	36	24.7
80 – 100 years	32	21.7	21	26.8
100 – 120 years	21	24.7	25	27.5
> 120 years	17	28.4	15	32.2
Uneven aged	12	21.4	3	29.9
All	215	20.0	140	24.9

Uneven-aged stands are generally relatively high, which indicates that most of these stands include full-grown trees, which have been included in the height assessments, probably combined with younger and smaller trees (of the same or a different species), which have not been included in the height assessment.

### ***Variation between the species groups***

The highest stands and dominant species were found for Beech, Spruce and to a lesser extent for Firs, European Oak and Remaining Broadleaves (Table 5.7). Relative low stands were found for the Remaining Conifers (based on the dominant stand height) and also for Alpine Conifers, Mediterranean Conifers and Evergreen Oaks (based on the height of the dominant species). Comparison between the Lorey's mean height and dominant height is possible for the species for which an (almost) equal number of stands is available. These results show that for most species, Lorey's dominant height is slightly larger than Lorey's mean height of the dominant species, like already shown in Fig. 5.10B. Unfortunately, the results in Table 5.7 for the species with large differences in height, are related to different sets of stands, and are therefore not directly comparable. Further analysis of these data, however, showed that large difference between Lorey's dominant stand height and Lorey's mean height of the dominant species were generally correlated with uneven-aged stand and with species which relatively often occurred in uneven-aged stand, such as Evergreen Oaks.

Table 5.7 Variation in the Lorey's mean height of the main tree species and the Lorey's dominant height of the entire stands as a function of the main tree species. Note that the mean height and dominant height are not comparable since different stands have been included.

Tree species cluster	Lorey's mean height of main species (m)			Lorey's dominant stand height (m)		
	N	Avg.	Std.	N	Avg.	Std.
Pine	41	17.7	3.2	27	20.0	5.5
Spruce	84	22.1	6.2	49	27.2	6.7
Firs	1	23.0		1	28.3	
Alpine Conifers	1	11.5		1	25.8	
Mediterranean conifers	19	12.9	5.0	2	24.6	0.3
Remaining Conifers	1	4.6		4	10.2	7.2
Beech	30	27.7	6.4	30	28.6	6.3
European Oak	13	21.3	5.2	14	26.3	4.0
Evergreen Oaks	12	6.1	1.3	2	18.6	0.1
Other Oaks	9	14.1	7.1	8	19.6	4.7
Remaining Broadleaves	4	33.7	31.7	2	26.5	8.1
All Conifers	147	19.5	6.4	84	24.0	7.6
All Broadleaves	68	21.2	12.2	56	26.3	6.3
All	215	20.0	8.7	140	24.9	7.2

### ***Variation within the major tree species***

Further investigation of the Lorey's dominant stand height as a function of the main tree species was only possible for the four major species, but not for the two Mediterranean species, due to insufficient data (0 and 2 stands for *Pinus pinaster* and *Quercus ilex*, respectively). The Lorey's dominant height of the stands dominated by *Picea abies* is generally larger than for the stands dominated by *Pinus sylvestris* (Fig. 5.11A). The values for *Pinus sylvestris* forests varied between 15 and 30 m, whereas the values for *Picea abies* forest varied between 17 and more than 40 m. The *Quercus robur/petraea* stands showed only little variation in Lorey's dominant height, with most values between 20 and 30 m. (Fig. 5.11B). The stands dominated by *Fagus sylvatica* mostly have a similar Lorey's dominant height, but 30% of the stand had values between 30 and 40 m.

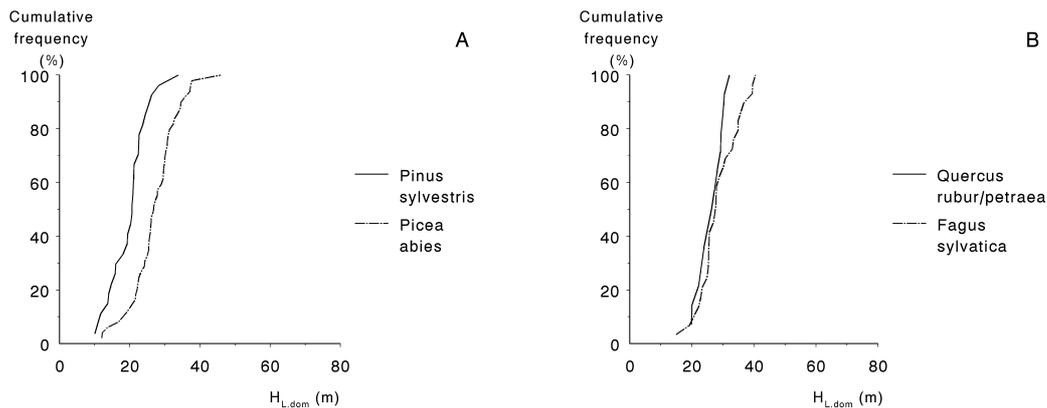


Figure 5.11 Lorey's dominant stand height at the Intensive Monitoring plots dominated by *Pinus sylvestris* and *Picea abies* (A), and by *Quercus robur/petraea* and *Fagus sylvatica* iilex (B) There were insufficient data to include *Pinus pinaster* and *Quercus ilex*..

The comparison of the mean height and mean diameter showed that most coniferous stand showed a strong relationship between these two variables. Most values were found in a range close to the relationship characterised by  $H_{L,lor}(m)=0.9 \cdot D_g(cm)$  (Fig. 5.12A). A large proportion of stands appeared to be larger at the same diameter (i.e. large, thin trees). It is known from literature, that above this limit of 0.9 the trees may become susceptible for storm damage. It has not been investigated what special conditions are the reason for the occurrence of stands with long, thin trees compared to short, thick trees (e.g. high altitude or longitude, high exposure to wind or a deviating management system).

Such as strong relationship between diameter and height was not found for the broadleaved species (Fig. 5.12B). The *Quercus ilex* stands were generally low, irrespectively of the observed diameters. Most stands with *Quercus robur/petraea* were generally close to a relationship of  $H_{L,lor}(m)=0.75 \cdot D_g(cm)$ , which indicates that the trees in these stands are generally shorter than most conifers when considering the same diameter. The stands with *Fagus sylvatica* showed a larger variation, with  $H_{L,lor}/D_g$  ratios between 0.25 and 1.00  $m \cdot cm^{-1}$ . This indicates that there might be considerable variations for these stands in the management system or the site conditions.

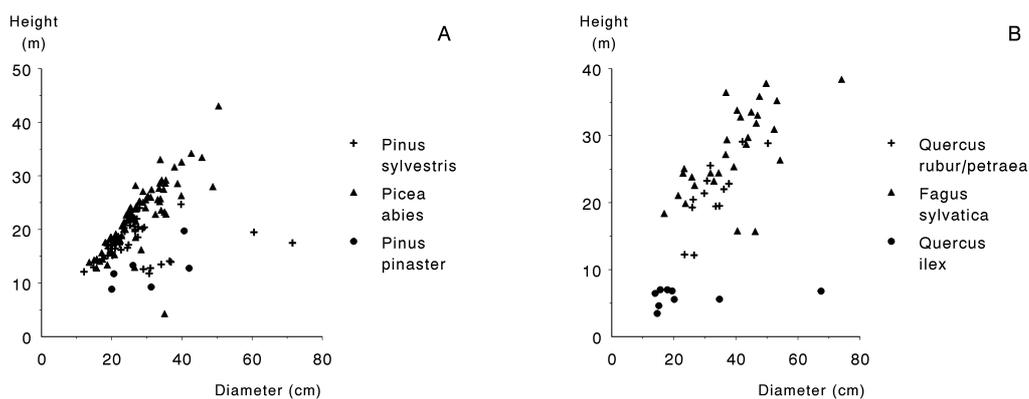


Figure 5.12 Comparison of Lorey's mean height with the quadratic mean diameter of the main tree species in stands with *Pinus sylvestris*, *Picea abies* and *Pinus pinaster* (A), and with *Quercus robur/petraea*, *Fagus sylvatica* and *Quercus ilex* (B).

## 5.4 Conclusions

The following conclusions can be drawn from the results discussed in the preceding sections:

- The available information on the measurements of stem diameters and tree heights offers good possibilities to derived relevant stand characteristics for about 65% of the 403 Intensive Monitoring Plots for which data until 1996 have been stored. At part of the plots, the evaluation was limited due to incomplete measurements (not all trees in a defined plot or subplot were measured) or by lack of methodological information. This requires further attention.
- The evaluated stands showed considerable variation in stand density, stand structure and stand height. The largest values were generally found for Spruce and Beech stands. Approximately 40% of the stands were complete monocultures, whereas the remaining stands were mostly only slightly mixed, except for many Oak stands. Most stands were relatively even-aged, except for the Evergreen oak stands. The contribution of secondary species in the basal area (biomass) is generally very low and they are also thinner and smaller than the dominant species, but they contribute considerably to the species diversity (especially by stem numbers) in mixed stands.



## 6 Atmospheric deposition

### 6.1 Introduction

Information on atmospheric deposition of nitrogen and sulphur compounds and of base cations is of key importance to understand the biogeochemical cycle in forest ecosystems. Elevated atmospheric deposition fluxes influence the soil solution chemistry and through that the nutritional status of forests. In the previous Technical Report (UN-ECE/EC; de Vries et al., 1998), it has already been made clear that nitrogen and sulphur deposition significantly influences the foliar N and S contents.

Atmospheric deposition can be distinguished in wet deposition or wash-out, where the particles and gases are extracted from the atmosphere by rain or snow and are deposited with the precipitation onto the underlying surfaces, and dry deposition, where particles (specifically base cations) and gases (specifically S and N compounds) deposit directly onto surfaces such as foliage and soils. Dry deposition may dominate closer to the source because particles gradually settle downward out of the plume onto the underlying surfaces. This deposition will occur in the absence of precipitation. At greater distances, the particles remaining in the plume are smaller and more able to stay aloft until precipitation occurs. Their relative importance further varies due to several environmental factors (See Section 6.2.4).

An indication of the wet deposition on forests can be derived from bulk deposition data nearby the forests, which also includes a fraction of dry deposition. Data on the differences in bulk deposition and wet deposition, using so called wet only samplers, however, show that this fraction is relatively small. Ivens (1990) found a mean fraction of 0.15 for  $\text{SO}_x$  and  $\text{NO}_x$ , 0.20 for  $\text{NH}_x$  and between approximately 0.20-0.30 for base cations, using data from Sweden, Czech Republic, Germany and the Netherlands.

Insight in the total deposition on a forest ecosystem, being the sum of wet and dry deposition is, however, more relevant as it gives information on the total atmospheric load, to be compared with e.g. a critical load. Total deposition can be derived directly by measuring both bulk (wet) precipitation and air quality. Multiplying the air concentrations by a so-called dry deposition velocity gives the dry deposition.

An other indication of the total (wet and dry) deposition on forest canopies can be derived from the sum of throughfall and stem flow (which equals the input to the soil system) corrected for canopy interaction, such as canopy uptake (in case of  $\text{NH}_4$ ) and canopy leaching (in case of base cations). The assumption is that throughfall (water dripping off the canopy onto the ground) and stem flow (water draining from the canopy to the ground along the stem structure of the vegetation) includes the contaminants previously deposited on forest canopy surfaces by dry deposition processes, since dissolution will take place as the precipitation interacts with the forest. Using this approach, one neglects dry deposition passing the forest canopy, but this effect is negligible, except for very open forests. For S and N compounds and base cations, those canopy interactions can be estimated from data on the bulk deposition, throughfall and stemflow of those compounds in combination with Na and Cl, as illustrated in this chapter. In case of heavy metals, one also needs litterfall data, but those compounds were not evaluated in this years report.

Important questions with respect to atmospheric deposition are:

- What is the relative contribution of wet and dry deposition in the potential acid input.
- What is the role of canopy uptake in modifying the nitrogen and base cation input fluxes.
- What is the relative contribution of N and S compounds in the potential acid input.
- What is the contribution of base cations in neutralising the acid input from the atmosphere.
- What is the order of magnitude of atmospheric inputs in view of critical loads.

Apart from various methodological aspects, described in Section 6.2, this chapter focuses on results related to those questions by (i) presenting methods and results of calculations of total (wet and dry) deposition accounting for canopy interactions (Section 6.3.1), (ii) ion ratios (specifically base cations or nitrogen versus sulphate) in bulk deposition, throughfall and total deposition (Section 6.3.2) and (iii) the range and geographic variation in atmospheric deposition distinguishing between bulk deposition, throughfall and total deposition (Section 6.3.3). Furthermore, attention is given to the impacts of site and stand characteristics on the atmospheric input, specifically total deposition (Section 6.3.4). Results on annual inputs are presented in  $\text{mol}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ . These results can be recalculated to  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  by a multiplication factor of 0.016 for S and 0.014 for N.

## 6.2 Methodological aspects

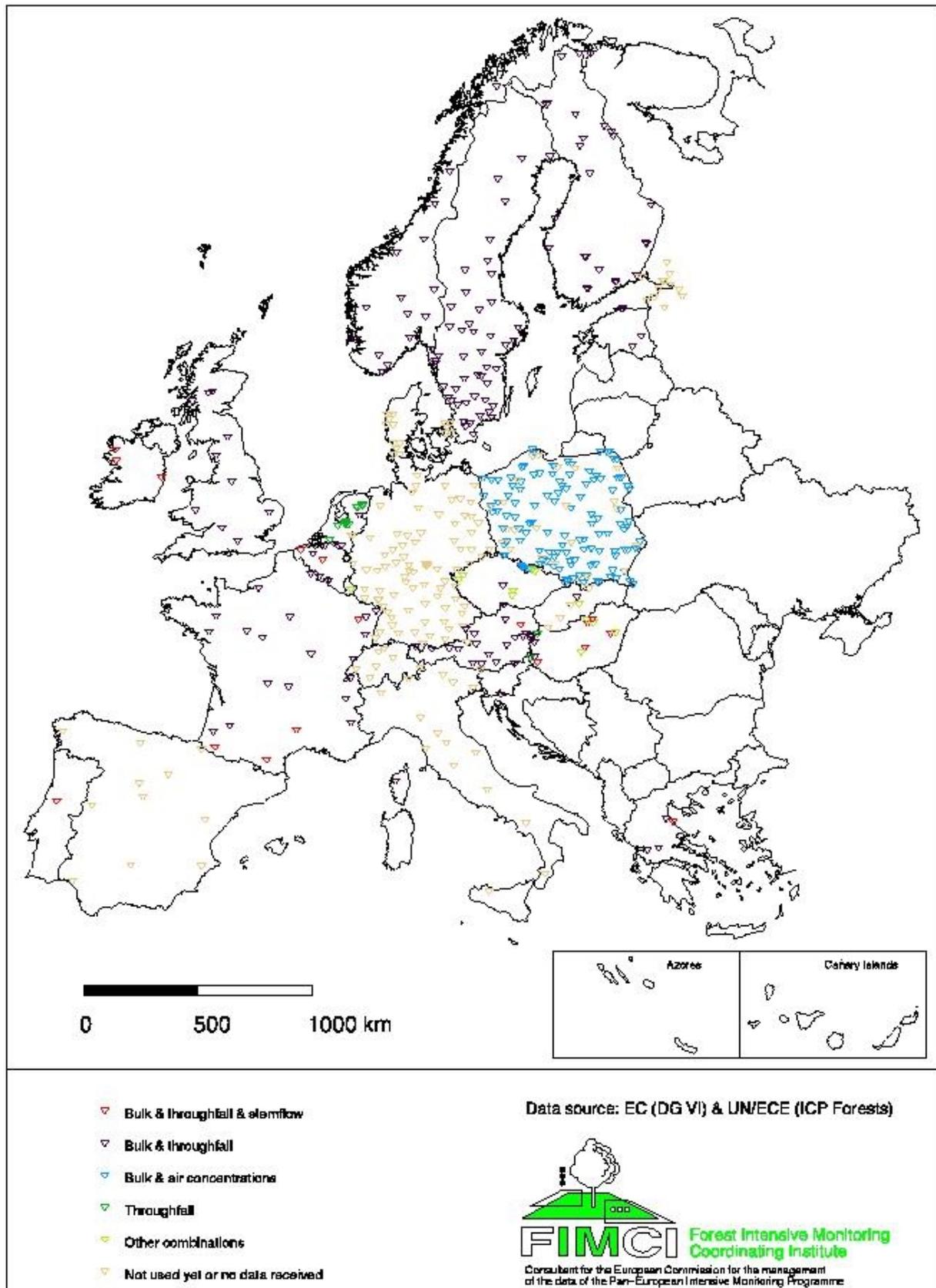
### 6.2.1 Locations

The total number of plots for which deposition measurements took place and consistent data were submitted in 1996 equalled 320 in a total of 18 countries. The total number of countries where deposition measurements took place equals 23 (see also Fig. 6.1) but some countries did not yet submit data for 1996 (Switzerland, Spain, Italy) whereas the data from two countries (Denmark, Germany) did not yet pass the validation process. Bulk deposition was measured at 301 plots in the same countries (Table 6.1).

With the exception of Poland, with a total number of 122 bulk deposition plots, throughfall was measured at all plots where bulk deposition was measured (198 plots). At a total of 19 plots (in the Netherlands and Hungary) throughfall was measured, whereas bulk deposition was not included. Stemflow was only measured at 23 plots in 8 different countries (Table 6.1).

The number of plots where the number of measurements was sufficient to compute annual deposition fluxes equalled 268 for bulk deposition, 163 for throughfall and 19 for stemflow (Table 6.1). At these plots, measurements are available for at least 75% of the year. At 144 plots annual fluxes for both bulk deposition and throughfall could be derived (at 19 of the 163 throughfall plots, no bulk deposition was measured).

The geographic variation of the plots where (i) bulk deposition, (ii) bulk deposition and throughfall and (iii) bulk deposition, throughfall and stemflow was measured in 1996 is given in (Fig 6.1). It shows that most deposition plots are concentrated in Central and Western Europe.



**Figure 6.1** Geographical distribution of the plots of the Pan-European Intensive Monitoring Programme and measurements made in the deposition survey of 1996

Table 6.1 Number of plots at which deposition, throughfall and stemflow was measured in 1996 in the various participating countries.

Country	Number of plots <sup>1)</sup>					
	Bulk deposition		Throughfall		Stemflow	
Finland	24	(19)	24	(19)	-	(-)
Sweden	48	(30)	48	(30)	-	(-)
Norway	17	(17)	17	(17)	-	(-)
U.K.	10	(10)	10	(10)	-	(-)
Ireland	3	(3)	3	(3)	3	(3)
Netherlands	2	(2)	14	(14)	-	(-)
Belgium	7	(6)	8	(6)	3	(3)
Luxembourg	1	(1)	1	(1)	1	(1)
France	25	(25)	25	(25)	4	(4)
Estonia	4	(-)	4	(-)	-	(-)
Poland	122	(122)	-	(-)	-	(-)
Czech Republic	1	(1)	1	(1)	-	(-)
Hungary	8	(7)	14	(12)	9	(7)
Austria	20	(20)	20	(20)	1	(1)
Croatia	1	(1)	1	(1)	-	(-)
Slovak Republic	3	(-)	3	(-)	-	(-)
Portugal	1	(1)	1	(1)	1	(-)
Greece	4	(4)	4	(3)	1	(-)
Total	301	268	198	163	23	19

<sup>1)</sup> Numbers refer to the data that were stored and used in the data evaluation. Numbers in brackets denote the plots for which annual deposition fluxes could be calculated.

## 6.2.2 Data assessment methods

Data assessment information for deposition monitoring until 1996 has been stored for 395 plots in 20 countries. For most monitoring plots, the methodological information was already submitted last year. Only a few changes were reported for the 1996 monitoring, whereas for 6 plots new, updated information has been received. The presentation of applied data assessment methods given below applies to the plots from which data have been used in the evaluations (320 plots; see also Section 6.1).

### *Sampling devices / sampling frequencies*

For monitoring of throughfall data use was mostly made of funnels (71%), whereas three different types of gutters were used at 9% of the plots. For the remaining 20%, no information was available. Funnels mostly had diameters ranging between 14-18 cm (67.4%), followed by funnels with a diameter of 20-20.5 cm (22.4%).

The sampling frequencies for throughfall measurements were mostly weekly or fortnightly. Only on 13% of the plots, for which sampling frequencies were reported, frequencies of 4-weekly or monthly periods were indicated. Sampling frequencies for stemflow were weekly at 8 of the 9 plots.

### *Number of samples*

To ensure that the determined atmospheric deposition data are representative for the monitoring plot, a good sampling set-up is essential. A sufficiently large number of samplers should be used

to get a representative average value, considering the spatial variation in throughfall. Generally recommended minimal requirements mentioned in literature are the installation of at least 10 randomly placed funnels with a diameter of 20 cm (Lövblad, 1994). For measuring throughfall the use of gutters is sometimes preferred above funnels (Draaijers et al., 1997), because they integrate the input over a larger canopy, thus yielding more representative estimates of the throughfall flux (e.g. Beier and Rasmussen, 1989).

The comparison between these recommendations in literature and actually used number and type of throughfall samplers in the Intensive Monitoring, showed that in general the numbers of samplers were sufficiently large (Fig. 6.2). On the plots where funnels are used (71% of all deposition plots) the number of throughfall samplers used mostly ranged from 10-15 (75%). 16-20 samplers were used on 17% of the plots where funnels are applied. On 4% of these plots more than 20 samplers are placed. Only on a few plots less than 10 deposition funnels were used. The minimum number of samplers per plot was reported to be 7. This was the case for only 4 % of the plots where funnels are used.

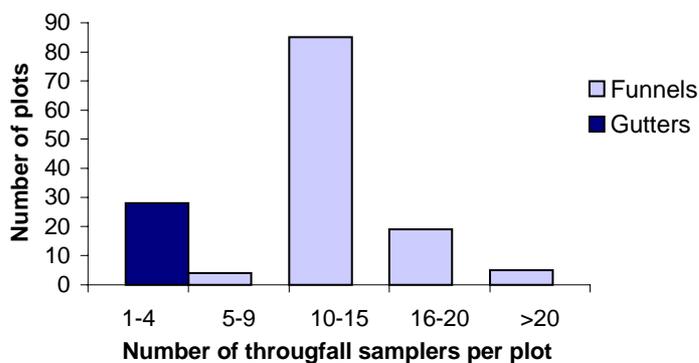


Figure 6.2 Number of samplers used to measure throughfall in 1996.

Representative sampling of stemflow is difficult, due to large variations between trees. It requires sampling of a large number of trees. A number of minimal 5-10 trees is mentioned in the literature (Lövblad, 1994). However, this number seems to be a rather conservative estimate (Draaijers et al., 1997). The number of 5 trees was not reached on 67 % of the plots. However, a sampling system in which multiple trees are connected to one collection tank, may have been reported as one sampler in the DAR-Q's. The number of sample trees may thus be underestimated to some extent.

In most cases bulk deposition was measured on one sampling site in the open field in the vicinity of the Intensive Monitoring plot. For 41.7% of all open field stations the reported distance was not longer than 1 kilometer, 6.6% being within 100 m, 13.9% between 100 and 500 m and 21.2% between 500 and 1000 m. No precise information was reported for the other 58.3% of the plots. The number of bulk deposition samplers per site ranged between 2 and 6 on the majority of the plots (97%) for which information was available. For 60% of the plots no information on numbers of bulk deposition samplers has been received.

### **Data comparability**

As discussed before, ringtests are essential when reliability and comparability of data are regarded. Insight in the quality of analysis by the various laboratories could be obtained from such

a ringtest carried out in 1996 (Lövsblad, 1997). A total of 133 laboratories from different countries participated in this exercise, of which 18 laboratories are also participating in the Intensive Monitoring programme. The performance of these 18 laboratories was compared to the whole group of 133 laboratories participating in the intercomparison exercise. The 18 laboratories in general had good results (5.9% outliers). In general no comparability problems for the deposition values existed. More information on the results is given in De Vries et al. (1998).

### 6.2.3 Data quality assurance

Procedures for quality assurance and quality control (QA/QC) included a check on all individual measurements with respect to:

- the balance between cations and anions
- the difference between measured and calculated electric conductivity
- the ratio between ion concentrations on an individual and an annual basis

In principle, those procedures should already be carried out by the laboratories producing the data, followed by a control by FIMCI during the data validation. The latter procedure showed that such a check was not always carried out by the individual laboratories. The balance between cations and anions, for example, showed that several countries submitted concentration data for sulphate, nitrate and ammonium in mg SO<sub>4</sub>.l<sup>-1</sup>, mg NO<sub>3</sub>.l<sup>-1</sup> and NH<sub>4</sub>.l<sup>-1</sup> instead of mg SO<sub>4</sub>-S.l<sup>-1</sup>, mg NO<sub>3</sub>-N.l<sup>-1</sup> and NH<sub>4</sub>-N.l<sup>-1</sup>. Furthermore, several measurements appeared to be contaminated by Na, as shown by extreme Na to Cl ratios, probably due to release from glass bottles (see also Annex 1).

The various procedures and results are described in detail below. The presented results are based on updates of submitted data, correcting the mistakes described above.

#### *Ionic balance*

On an equivalent basis, the sum of all major cations should equal the sum of all major anions. The percentage difference was therefore calculated according to:

$$PD = 100 * \frac{(\text{cat} - \text{an})}{0.5 * (\text{cat} + \text{an})} \quad (6.1)$$

$$\text{cat} = [\text{Ca}^{++}] + [\text{Mg}^{++}] + [\text{Na}^+] + [\text{K}^+] + [\text{H}^+] + [\text{NH}_4^+] \quad (6.2)$$

$$\text{an} = \text{Alk} + [\text{SO}_4^{--}] + [\text{NO}_3^-] + [\text{Cl}^-] \quad (6.3)$$

where:

PD = percentage difference (%)

Alk = alkalinity (mmol<sub>c</sub>.m<sup>-3</sup>)

The basic assumption is that the charge of the other cations and anions present in solution can be neglected. In the case of throughfall and stemflow, the presence of organic acids, indicated by concentrations of DOC, do, however, cause differences leading to a cation excess. When DOC is

measured, an empirical correction for the charge of those organic anions (e.g. Oliver et al., 1983) is possible. This has not yet been done, since it is considered inadequate when DOC concentrations are high (Ulrich et al., 1998). In bulk deposition, the concentrations of low molecular organic acids, such as formic and acetic acid, only have a minor role in the ionic balance.

Figure 6.3, shows the relationship between the sum of cations and anions in bulk deposition and throughfall using all the individual measurements at all Intensive Monitoring plots. The checks were limited to those measurements where all major ions mentioned in Eq. (6.2) and Eq. (6.3) were available. Only alkalinity was allowed to be missing in plots where the pH is less than 4.5. In that case the alkalinity, which stands for the sum of  $\text{HCO}_3^-$ ,  $\text{CO}_3^{2-}$  and  $\text{OH}^-$  corrected for  $\text{H}^+$ , was assumed to be negligible (see section on ionic ratios). This included approximately 35-45% of the measurements (See also Table 6.3).

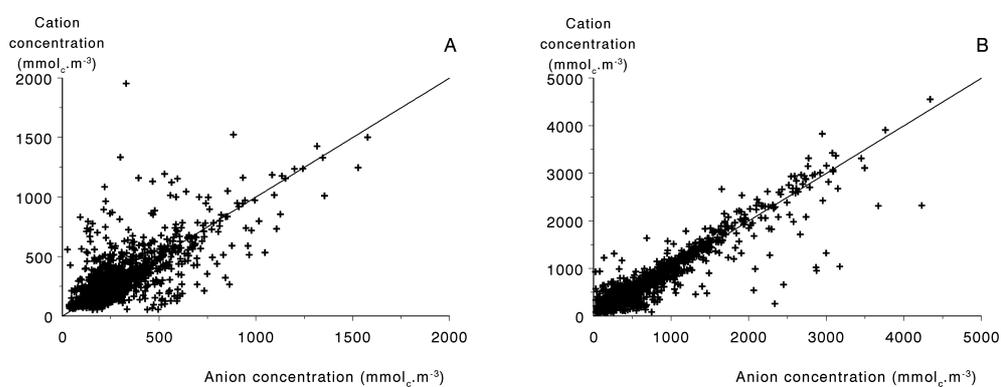


Figure 6.3 Relationships between the sum of cations and the sum of anions in bulk deposition (A) and throughfall (B), using all individual measurements at all Intensive Monitoring plots. Beware of the difference in axes between bulk deposition and throughfall. The solid line represents the 1:1 line.

The results show large differences varying from a strong anion excess to a large cation excess. Comparable results were obtained for stemflow, for which the number of measurements is much less. This implies that the results are far from satisfactory. In general, it is required that PD is less than 10% for bulk deposition and less than 20% for throughfall when the sum of cations and anions is larger than  $500 \text{ mmol}_c.\text{m}^{-3}$  (WMO, 1992; Ulrich et al., 1998). Larger relative differences are acceptable at low concentrations of the sum of cations and anions (Table 6.2).

Table 6.2 The required criteria for the ionic balance (WMO, 1992).

Cations+anions ( $\text{mmol}_c.\text{m}^{-3}$ )	Acceptable difference (%)
$\leq 50$	$\leq 60$
50 – 100	$\leq 30$
100 – 500	$\leq 15$
$> 500$	$\leq 10$

Approximately 50-60% of the measurements only appears to fulfil this requirement (Table 6.3).

Table 6.3 Percentage of measurements in different ranges for the relative difference in sum of cations and sum of anions.

Difference (%)	Percentage of measurements <sup>1)</sup>		
	Bulk deposition <sup>2)</sup>	Throughfall <sup>3)</sup>	Stem flow <sup>4)</sup>
< -30	16.3	9.7	12.4
-30 - -20	5.1	4.6	5.8
-20 - -10	5.6	5.8	7.1
-10 - 0	13.2	12.4	17.3
0 - 10	17.0	24.1	21.8
10 - 20	12.7	16.0	13.8
20 - 30	9.5	8.4	8.9
> 30	20.6	19.0	12.9

<sup>1)</sup> This includes measurements where all ions including alkalinity have been measured or alkalinity has not been measured but pH is less than 4.5.

<sup>2)</sup> The number of complete measurements equals 1549 (total number of measurements is 4750)

<sup>3)</sup> The number of complete measurements equals 1734 (total number of measurements is 3600)

<sup>4)</sup> The number of complete measurements equals 225 (total number of measurements is 534)

Results indeed showed that the percentage difference between cations and anions decreased with an increase in ionic concentrations, especially in throughfall. However, even at very low ionic strength (concentrations of cations and anions  $< 100 \text{ mmol}_c \cdot \text{m}^{-3}$ ), the differences were often still larger than the acceptable differences of 30-60% (Fig. 6.4).

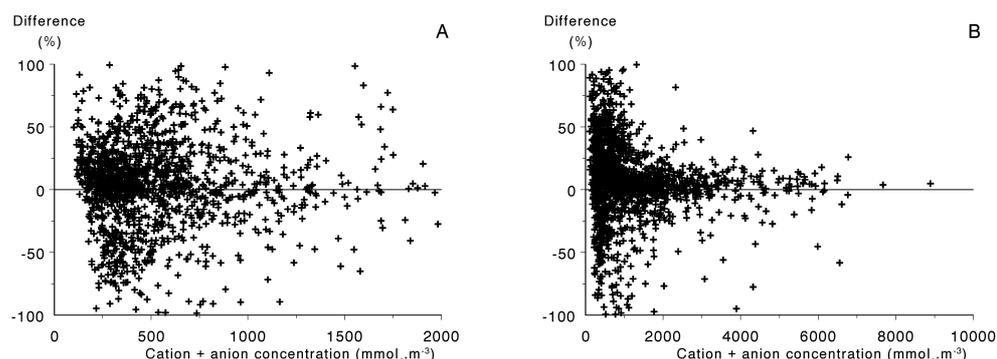


Figure 6.4 The percentage difference between cations and anions as a function of the sum of the concentrations of cations and anions in the bulk deposition (A) and throughfall (B).

The imbalance may partly be due to inaccuracies in less relevant ions, such as Na and Cl (see later). In general, however, there was no clear relationship between the difference in cations and anions and the Na to Cl ratio. Only at very high Na to Cl ratios (above 30), the sum of cations was consistently much higher than the sum of anions. This result poses questions to the quality of the data. Because of these questions, interpretations of results have been made carefully in view of its plausibility in the light of available literature. Because of the large differences in cations and anions in both bulk deposition and throughfall, the calculation of total deposition and of canopy interactions based on those fluxes has also been simplified (Section 6.2.4).

### Electric conductivity

Another quality check is the difference between measured and calculated electric conductivity, which should be less than 20% when the measured conductivity is larger than  $30 \mu\text{S} \cdot \text{cm}^{-1}$  (WMO, 1992; Ulrich et al., 1998). Electric conductivity (EC) is a measurement of the ability of an

aqueous solution to carry an electric current. Apart from temperature, this ability depends on the type and concentration (activity) of ions in solution according to:

$$EC = \sum_i \lambda_i \cdot f_i \cdot c_i \quad (6.4)$$

where:

EC = electric conductivity ( $\mu\text{S}\cdot\text{cm}^{-1}$ )

$\lambda_i$  = equivalent ionic conductance, being the capacity of a single ion to carry an electric current in ideal conditions of infinite solution at 20°C ( $\text{kS}\cdot\text{cm}^2\cdot\text{eq}^{-1}$ )

$c_i$  = concentration of ion i with i = H, Ca, Mg, K, Na,  $\text{NH}_4$ ,  $\text{NO}_3$ ,  $\text{SO}_4$ , Cl, Alk ( $\text{mmol}\cdot\text{m}^{-3}$ )

$f_i$  = activity coefficient of ion i.

Values of  $\lambda_i$  for the various ions, together with the equivalent weight by which the concentrations (submitted in  $\mu\text{g}\cdot\text{l}^{-1}$ ) are divided (to get  $\text{mmol}\cdot\text{m}^{-3}$ ) are given in Table 6.4.

Table 6.4 Equivalent ionic conductance at 20°C and equivalent weights for the various considered ions in deposition.

Element	Equivalent ionic conductance ( $\text{kS}\cdot\text{cm}^2\cdot\text{eq}^{-1}$ )	equivalent weight ( $\text{g}\cdot\text{eq}^{-1}$ )
H	0.3151	1
Ca	0.0543	20
Mg	0.0486	12
K	0.0670	39
Na	0.0459	23
$\text{NH}_4\text{-N}$	0.0670	14
$\text{NO}_3\text{-N}$	0.0636	14
$\text{SO}_4\text{-S}$	0.0712	16
Cl	0.0680	35.5
Alk	0.0394	1

The EC values of each individual measurement were calculated using Eq. (6.4) and using the values given in Table 6.4. Even though DOC is generally not measured, which unbalances the difference between cations and anions (see before), this does not have an important influence on the calculated conductivity. Activity coefficients were calculated as a function of the ionic strength (I), using the Davies equation (Stumm and Morgan, 1981). Results of the calculation in comparison to measured values are given in Figure 6.5.

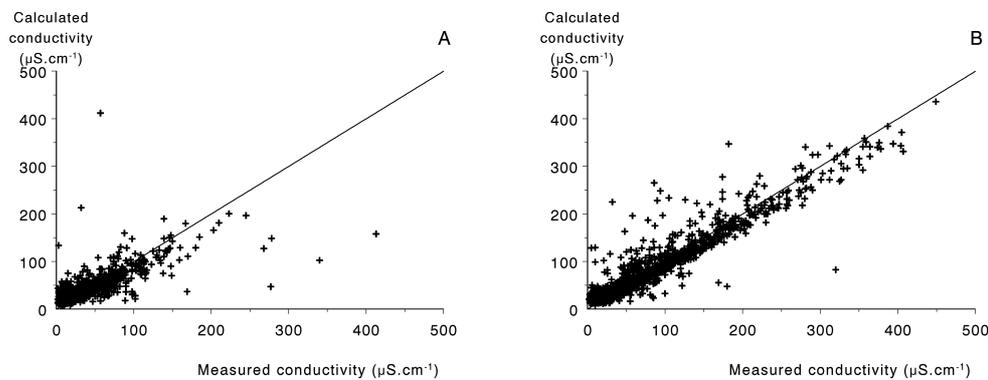


Figure 6.5 Relationship between the calculated and measured conductivity in bulk deposition (A) and throughfall (B) using all individual measurements at all Intensive Monitoring plots. The solid line represents the 1:1 line.

Results appear to be slightly better than the difference between the sum of cations and anions. In many cases there are, however, large differences between the measured and calculated conductivity in bulk deposition (Fig. 6.5A), throughfall (Fig. 6.5B) and stemflow (not shown). This can also be derived from the percentage difference between calculated and measured conductivity:

$$PD = 100 * \frac{(EC_{\text{calc}} - EC_{\text{meas}})}{EC_{\text{calc}}} \quad (6.5)$$

According to WMO (1992), the discrepancy between measured and calculated conductivity should therefore be no more than 20% at a measured conductivity above 30  $\mu\text{S}\cdot\text{cm}^{-1}$ . At low ionic strength, the acceptable differences are higher, a maximum difference of 30% is generally required.

Results (Table 6.5) show that this requirement is fulfilled by a majority of the measurements. The percentage of measurements within a difference of  $\pm 20\%$  increased from approximately 55% for bulk deposition to 65% for throughfall and 75% for stemflow (Table 6.5). Nevertheless, a large percentage of the measurements does not fulfil the requirements. A much stricter quality control on the measurements by the various laboratories (and NFC's) before submitting the data thus appears to be crucial.

*Table 6.5 Percentage of measurements in different ranges for the difference between calculated and measured conductivity.*

Difference (%)	Percentage of measurements		
	Bulk deposition	Throughfall	Stem flow
< -30	4.4	3.1	0.9
-30 - -20	4.0	3.9	1.8
-20 - -10	12.9	12.9	13.4
-10 - 0	18.9	26.0	33.5
0 - 10	12.6	14.9	22.8
10 - 20	9.2	7.7	4.0
20 - 30	5.7	6.1	5.8
> 30	32.3	25.4	17.9

### ***Ionic ratios***

The correlation between ions in solution and the covariance between ion concentration ratios is a third possibility to check the quality of the data. An important check is the ratio between Na and Cl. Assuming that seasalt is a dominant source of both ions, the Na to Cl ratio should resemble the ion ratio in seawater being equal to 0.858  $\text{eq}\cdot\text{eq}^{-1}$ . Ivens (1990) found a Na to Cl ion ratio mostly varying between 0.7 and 1.0 in annual bulk deposition and throughfall fluxes with a median value resembling the ratio in seawater (0.84 in bulk deposition and 0.88 in throughfall). Draaijers (pers. comm.) stated that on an annual basis, the Na to Cl ratio should vary between 0.5 and 1.0. At the Intensive Monitoring plots, the ratios in Na and Cl concentration showed a wider variation, specifically in the individual measurements (Fig. 6.6) but also on an annual basis (Fig 6.7).

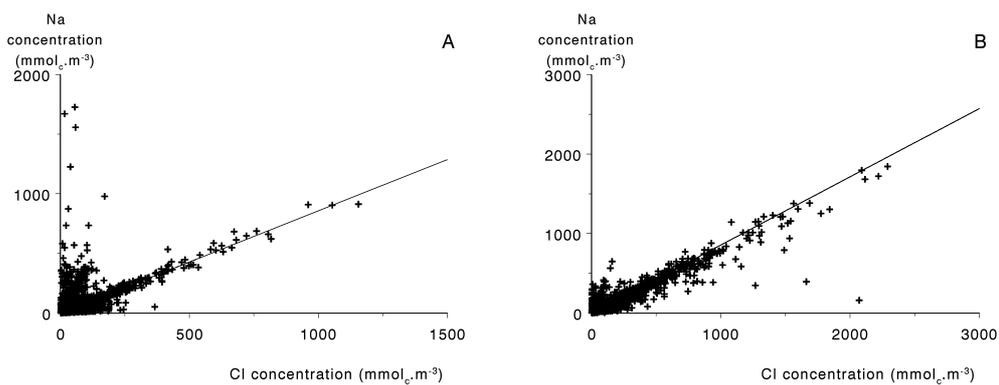


Figure 6.6 Relationships between the concentrations of Na and Cl in individual measurements of bulk deposition (A) and throughfall (B). The solid line represents the Na/Cl ratio in sea water ( $0.858 \text{ eq.eq}^{-1}$ ). Beware of the difference in axes between bulk deposition and throughfall. The solid line represents the 1:1 line.

In general, ratios stronger resembled seawater at higher concentrations. Furthermore, results were better for throughfall than for bulk deposition. Especially at low Cl concentrations (and fluxes), there were quite a lot of measurements with high Na concentrations. In those situations, extremely high Na/Cl ratios were found, exceeding values of 10 up to 100. Such ratios do indicate Na contamination. The earlier mentioned relationship between a consistent cation excess and extremely high Na/Cl ratios confirms this indication. Application of linear regression analyses without an intercept, while using the individual measurements, resulted in a slope (being the ‘average’ Na/Cl ratios) of 0.64 for bulk deposition ( $R^2_{\text{adj}} = 20\%$ ) and of 0.75 for throughfall ( $R^2_{\text{adj}} = 89\%$ ). For the annual fluxes the slopes equalled 0.76 in bulk deposition ( $R^2_{\text{adj}} = 76\%$ ) and 0.80 in throughfall ( $R^2_{\text{adj}} = 94\%$ ).

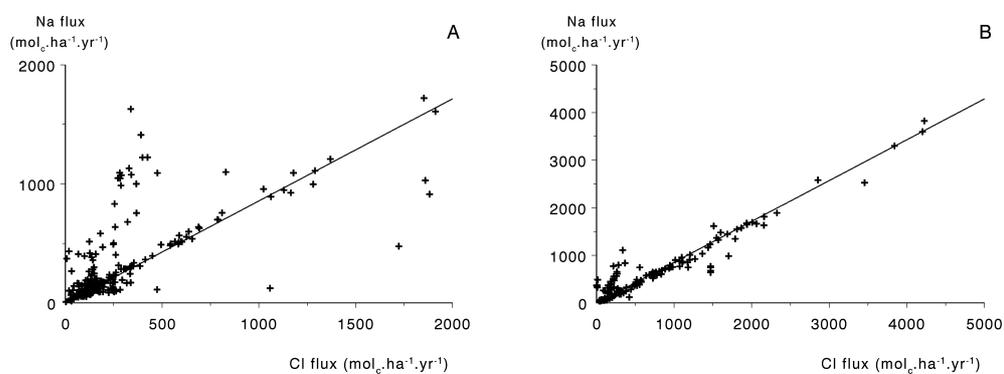


Figure 6.7 Relationships between the annual fluxes of Na and Cl in bulk deposition (A) and throughfall (B). The solid line represents the Na/Cl ratio in sea water ( $0.858 \text{ eq.eq}^{-1}$ ). Beware of the difference in axes between bulk deposition and throughfall. The solid line represents the 1:1 line.

Actually, wider ranges were specifically observed in Central and Southern Europe, where the influence of seasalt on the Na and Cl deposition is likely to be less. In the countries with a larger influence of the sea, the Na to Cl ratio in all individual measurements generally varied between 0.5 and 1.5. The median values in those countries varied mostly between 0.8 and  $1.0 \text{ eq.eq}^{-1}$ , strongly resembling the ionic ratio in sea water ( $0.858 \text{ eq.eq}^{-1}$ ) (Table 6.6).

Table 6.6 Ranges between which 90% of the Na to Cl ratios (eq.eq<sup>-1</sup>) varied in bulk deposition and throughfall as a function of country.

Country	Na/Cl ratio (eq.eq <sup>-1</sup> )					
	Bulk deposition			Throughfall		
	5%	50%	95%	5%	50%	95%
Finland	0.59	1.03	2.32	0.44	0.88	1.54
Sweden	0.62	0.96	1.54	0.51	0.81	1.24
Norway	0.62	0.93	1.54	0.51	0.82	1.16
U.K.	0.68	0.89	1.23	0.59	0.80	1.19
Ireland	0.36	0.83	1.03	0.66	0.85	1.35
Netherlands	0.60	0.77	1.02	0.54	0.82	11.27
Belgium	0.63	0.96	1.64	0.42	0.82	1.31
Luxembourg	0.39	0.81	1.01	0.51	0.64	0.95
France	0.42	0.89	1.23	0.38	0.75	1.18
Poland	0.26	0.93	6.37	-	-	-
Czech Republic	0.10	0.21	1.07	0.15	0.27	0.68
Hungary	0.83	2.16	17.75	0.57	2.21	9.11
Austria	0.48	2.64	13.10	0.30	1.79	10.66
Slovak Republic	0.21	0.80	5.85	0.28	0.77	4.11
Portugal	0.82	1.51	2.10	0.78	1.08	2.80
Greece	0.14	0.35	0.71	0.22	0.48	0.85
All	0.31	0.94	4.36	0.40	0.83	3.53

Another check is the relationship between alkalinity and pH. In general, alkalinity should increase with pH, since there is an equilibrium between HCO<sub>3</sub><sup>-</sup> and CO<sub>3</sub><sup>2-</sup> with H<sup>+</sup>, which is mainly determined by the CO<sub>2</sub> pressure. Results indeed showed such an increase, even though low alkalinity values were also observed at high pH values (Fig. 6.8). One measurement in throughfall with a very high alkalinity (above 2000 mmol<sub>c</sub>.m<sup>-3</sup>) at a pH of 4.5 is clearly an outlier (Fig. 6.8B). Positive alkalinities below pH 4.5, as sometimes observed, are very unlikely. Inversely, the results show clearly that alkalinity can generally not be neglected at pH values above 4.5.

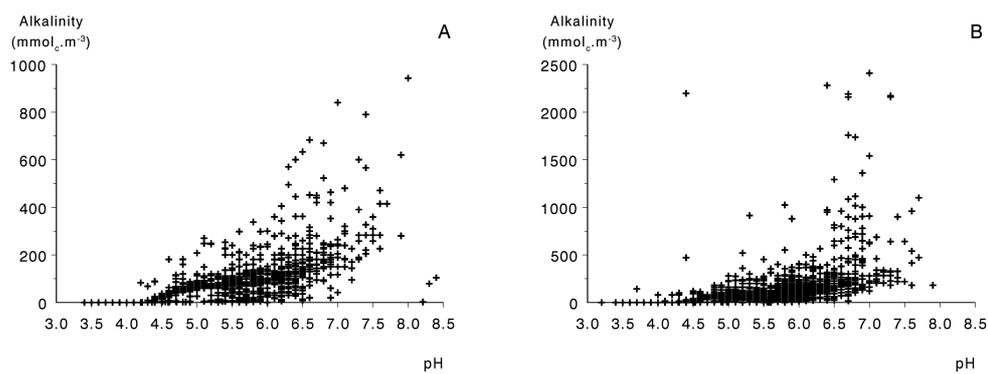


Figure 6.8 Relationships between alkalinity and pH in bulk deposition (A) and throughfall (B). Beware of the difference in axes between bulk deposition and throughfall.

## 6.2.4 Data evaluation methods

### *Calculation of annual fluxes of bulk deposition, throughfall and stemflow*

Annual deposition fluxes of the major ions were calculated by multiplying the biweekly or monthly precipitation amounts (in mm) by the ion concentrations ( $\text{meq}\cdot\text{m}^{-3}$ ) and summing up to one year. With respect to stemflow, it was assumed that the submitted data on mm of stemflow water were correctly related to the total area of the plot. One approach to do this is to multiply the amount of stemflow (in mm) at the sampled trees with the ratio of the basal area of the stand ( $\text{m}^2$ ) divided by the basal area of the sampled trees ( $\text{m}^2$ ) and subsequently divide it by 10000 (to get the input in mm per ha). Some countries supplied deposition data for only part of the year. In these cases, the total flux and average concentration for the entire year were computed by assuming that the deposition in the period without data was equal to the average deposition in the rest of the year. This adjustment was only made if measurements were available for at least 75 % of the year; otherwise the data were not used.

Stemflow was only measured at a limited number of plots (Section 6.2.1). At plots where such information was missing, the annual stemflow was estimated from the annual throughfall according to (Ivens, 1990)

$$X_{\text{sf}} = X_{\text{tf}} \cdot \alpha / (1 - \alpha) \quad (6.6)$$

where:

X = a given ion (H, Ca, Mg, K, Na,  $\text{NH}_4$ ,  $\text{NO}_3$ ,  $\text{SO}_4$ , Cl)  
sf = stemflow ( $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ )  
tf = throughfall ( $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ )  
 $\alpha$  = an empirical value

For coniferous forests, the value of  $\alpha$  was calculated as a function of stand age according to (Ivens, 1990):

$$\begin{aligned} \alpha &= 0.24 && \text{age} < 20 \\ \alpha &= 0.31 - 0.0034 \cdot \text{age} && 20 < \text{age} < 90 \\ \alpha &= 0.0 && \text{age} > 90 \end{aligned} \quad (6.7)$$

Actually, Ivens (1990) assumed that  $\alpha$  was twice as high for H than for the other ions (Ca, Mg, K, Na,  $\text{NH}_4$ ,  $\text{NO}_3$ ,  $\text{SO}_4$  and Cl), but a comparison of calculated and measured stemflow of 19 plots (see Section 6.3.1) did not substantiate this assumption. For deciduous forests,  $\alpha$  was set at 0.12 independent of age. The same values were used for coniferous forests with an unknown age (Ivens, 1990).

### *Calculation of total deposition from bulk deposition, throughfall data and stemflow*

Total deposition can be derived by correcting the input by both throughfall and stemflow for exchange processes occurring at the forest canopy. Canopy exchange processes are influenced by the input of ions from the atmosphere, the foliar nutrient status, the age distribution of needles, abiotic stress like drought and temperature extremes and by biotic stresses, such as insect plagues (Draaijers et al., 1998).

One method to calculate total deposition from bulk deposition, throughfall and stemflow data is the canopy budget model developed by Ulrich (1983) which was extended by Bredemeier (1988) and Van der Maas et al. (1991). A so-called ‘filtering approach’, which is part of this budget model, was used to calculate total deposition fluxes of base cations according to (Ulrich, 1983):

$$BC_{td} = \frac{Na_{tf} + Na_{sf}}{Na_{bd}} \cdot BC_{bd} \quad (6.8)$$

where:

BC = Ca, Mg, K

td = total deposition ( $\text{mol}_c \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ )

bd = bulk deposition ( $\text{mol}_c \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ )

Eq. (6.8) is based on the assumption that (i) Na does not interact with the forest canopy (tracer) and (ii) the ratios of total deposition over bulk deposition are similar for Ca, Mg, K and Na. Canopy leaching induced by the internal cycle of these nutrients, was thus computed by the difference between the sum of BC in throughfall and stemflow minus total deposition according to:

$$BC_{ce} = BC_{tf} + BC_{sf} - BC_{td} \quad (6.9)$$

where:

ce = canopy exchange ( $\text{mol}_c \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ )

Canopy exchange of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  was assumed negligible. The  $\text{NH}_4$  throughfall and stemflow flux was corrected for canopy uptake to calculate the total deposition of  $\text{NH}_4$  according to (Van der Maas et al., 1991; Draayers et al., 1994):

$$\text{NH}_{4,td} = \text{NH}_{4,tf} + \text{NH}_{4,sf} + \text{NH}_{4,ce} \quad (6.10)$$

with:

$$\text{NH}_{4,ce} = \left( \frac{\text{NH}_{4,tf}}{\text{NH}_{4,tf} + \text{H}_{tf} \cdot xH} \right) \cdot BC_{ce} \quad (6.11)$$

where:

xH = an efficiency factor of H in comparison to  $\text{NH}_4$

Eq. (6.11) is based on the assumption that total canopy uptake of  $\text{H}^+$  and  $\text{NH}_4^+$  is equal to the total canopy leaching of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{K}^+$  taking place through ion exchange. Based on experiments in the laboratory (Van der Maas et al., 1991), it is assumed that  $\text{H}^+$  has per mol an exchange capacity six times larger than  $\text{NH}_4^+$  ( $xH = 6$ ). Actually, Draayers et al. (1998) corrected the base cation leaching in Eq. (6.11) by subtracting the leaching of weak acids. This was, however, not included in our calculations since the estimation of the weak acid concentration, based on either (i) the sum of alkalinity and  $\text{RCOO}^-$  derived from DOC or (ii) the difference in concentration of cations minus strong acid anions ( $\text{SO}_4$ ,  $\text{NO}_3$ , Cl), was considered inadequate (Section 6.2.3). The total deposition of protons was calculated as:

$$H_{td} = H_{tf} + H_{sf} + H_{ce} \quad (6.12)$$

with:

$$H_{ce} = BC_{ce} - NH_{4, ce} \quad (6.13)$$

A more detailed description of the model is presented by Draaijers et al. (1994) and Draaijers and Erisman (1995). Up to now, several basic assumptions in the model (e.g. the ratio in exchange capacity between  $H^+$  and  $NH_4^+$ ) are not properly evaluated for different environmental conditions (tree species, ecological setting, pollution climate) which limits its application (Draaijers et al., 1994; Draaijers and Erisman, 1995). Furthermore, the model has only been validated in relatively polluted areas, such as the Netherlands and Denmark.

An estimate for canopy uptake of oxidised and reduced nitrogen was also obtained by using empirical results of the Integrated Forest Study (IFS) reported by Johnson and Lindberg (1992). For 12 sites in the USA, Johnson and Lindberg (1992) found that the above ground exchange (uptake) of inorganic nitrogen,  $N_{ce}$ , was significantly related ( $r^2 = 0.66$ ) to the total deposition of inorganic nitrogen,  $N_{td}$ , according to:

$$N_{ce} = 0.41 \cdot N_{td} + 54.2 \quad (6.14)$$

Eq. (6.14) was based on a significant relationship ( $r^2 = 0.80$ ) between the throughfall and stemflow flux of inorganic nitrogen,  $N_{tf} + N_{sf}$ , and the independently measured total deposition,  $N_{td}$ , according to:

$$N_{tf} + N_{sf} = 0.59 \cdot N_{td} - 54.2 \quad (6.15)$$

Combining equation (6.14) and (6.15) provides the relationship between the canopy exchange and the throughfall + stemflow flux of inorganic nitrogen (in  $eq \cdot ha^{-1} \cdot yr^{-1}$ ):

$$N_{ce} = 0.69 \cdot (N_{tf} + N_{sf}) + 91.9 \quad (6.16)$$

Johnson and Lindberg (1992) made their measurements at sites situated in areas with relatively low air concentrations of N compounds in comparison to those found in certain areas in Europe. Throughfall and stemflow fluxes of inorganic nitrogen in the IFS ranged between 100 and 1000  $eq \cdot ha^{-1} \cdot yr^{-1}$ . Equation (6.16) therefore can only be applied for this range of inorganic nitrogen fluxes. In this range, the canopy uptake ranges between 150 and 780  $mol_c \cdot ha^{-1} \cdot yr^{-1}$ , thus implying a total deposition of 250 to 1780  $mol_c \cdot ha^{-1} \cdot yr^{-1}$ . It is, however, unlikely that uptake rates will increase linearly at higher throughfall and stemflow fluxes because nitrogen saturation in the canopy might be expected. An estimated N uptake of 600  $mol_c \cdot ha^{-1} \cdot yr^{-1}$  at the Solling Spruce site (Eilers et al., 1997), receiving a high deposition of N (approximately 3000  $mol_c \cdot ha^{-1} \cdot yr^{-1}$ ), is an indication for this. Another restriction for using equation (6.16) is that it only applies to spruce and spruce-fir forests. Other tree species showed a rather constant inorganic N uptake of 200-300  $mol_c \cdot ha^{-1} \cdot yr^{-1}$  with only little response to deposition amount (Johnson and Lindberg, 1992). Ivens (1990) also suggested an above ground inorganic N uptake of 150-350  $mol_c \cdot ha^{-1} \cdot yr^{-1}$ .

In calculating the canopy uptake of NH<sub>4</sub> for spruce/fir forests we took the minimum of Eq. (6.11) and Eq. (6.16) with an absolute minimum and maximum of 100 and 800 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>, respectively (see Eq. 6.16). For all other forests, Eq. (6.11) was used to calculate the NH<sub>4</sub> canopy uptake with an absolute minimum and maximum of 100 and 400 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>, respectively.

***Assessments of relationships between atmospheric deposition and stand / site characteristics***

The extent or efficiency of dry deposition will depend upon features such as topography and surface roughness. For example, upslope and hilltop positions may receive relatively more dry deposition (and cloud water deposition) because the landscape effectively intersects the airborne plume. As the air mass containing the contaminants passes, the particles become impinged on the elevated surfaces. Clearly, surface roughness is important: tree canopies will intersect and entrain more airborne particles than adjacent low vegetation canopies such as grass. The process of wet deposition is less dependent on the elevation and features of the underlying surface. None the less, the elevation even affects the fate of the contaminants washed out of the plume leading to wet deposition, by so-called seeder feeder processes.

An overview of the expected relationships between stand and site characteristics and atmospheric deposition has been given in Section 3.3.1 (see also Fig. 3.1). More specifically, region (differences in air pollution levels) and altitude, the throughfall (total deposition) flux is largely influenced by the tree species (e.g. Draayers et al., 1992), tree height (e.g. Stevens, 1987) and canopy coverage (e.g. Draayers et al., 1992) which all affect the surface roughness. Canopy coverage is in turn related to indices for stand density and stand structure. When using throughfall data, soil type may also influence the results of base cations, as it affects the internal cycle in the ecosystem of these elements. The influence of stand and site characteristics on the selected key parameters for atmospheric deposition was tested by means of ordination techniques, viz. a principal component analyses (Section 3.3.2) and multiple regression approaches (Section 3.3.3). The regression models were of the following general type:

$$\begin{aligned} \log(\text{key parameter}) = & f_1(\text{tree species}) + f_2(\text{climatic region}) + f_3(\text{altitude}) + \\ & f_4(\text{stand height}) + f_5(\text{stand density index}) + \\ & f_6(\text{stand structure index}) + f_7(\text{precipitation}) \end{aligned} \quad (6.17)$$

The influence of each characteristic on a given key parameter was thus assessed while accounting for the impact of the other relevant stand and site characteristics. The approach implies that a stratification is made with respect to tree species and climate region characteristics. The logarithmic transformation was performed because most parameters had a skewed log-normal distribution. The key parameters used were the measured throughfall fluxes of SO<sub>4</sub>, NO<sub>3</sub> and NH<sub>4</sub> and the calculated total deposition fluxes of Ca, Mg and K. With respect to SO<sub>4</sub>, NO<sub>3</sub> and NH<sub>4</sub>, throughfall was assumed to be representative for total deposition. In case of NH<sub>4</sub>, the calculated total deposition was also used (see above). In case of ordination, the six parameters were simultaneously related to the mentioned predictor variables.

Actually, the available information on stand height, stand density index and stand structure index was very limited at the plots where atmospheric deposition was measured (35 plots). Consequently, both analyses were also performed at the total number of 125 plots without using these predictor variables.

## 6.3 Results and discussion

### 6.3.1 Relationships between bulk deposition, throughfall and total deposition

#### *Sulphur and nitrogen compounds*

A first indication of the relative importance of wet and dry deposition of S and N compounds can be derived from the relationship between measured annual throughfall and bulk deposition at the same plots. Results show that the difference between the input in throughfall and in bulk deposition was generally higher for  $\text{SO}_4$  than for the N compounds especially at low N deposition (Fig. 6.9).

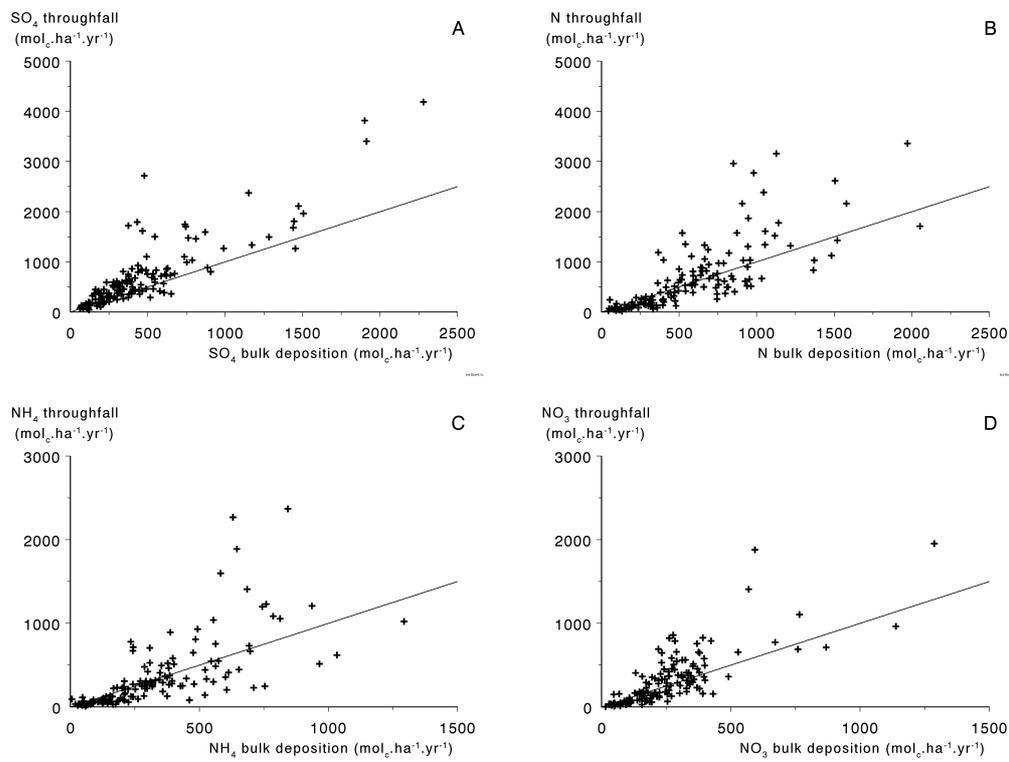


Figure 6.9 Relationships between the annual input by throughfall and bulk deposition for  $\text{SO}_4$  (A), N (B),  $\text{NH}_4$  (C) and  $\text{NO}_3$  (D) at 144 Intensive Monitoring Plots. The solid line represents the 1:1 line.

This can also be illustrated by (i) the range in throughfall to bulk deposition ratios for the various elements and (ii) the regression coefficients that were derived from a simple linear regression between throughfall and bulk deposition (Table 6.7).

Higher throughfall to bulk deposition ratios for  $\text{SO}_4$  compared to N compounds can partly be due to a higher dry deposition velocity for  $\text{SO}_2$  than for  $\text{NO}_x$  and  $\text{NH}_x$  or to higher local sources of S compared to N. The large number of plots (>50%) with a throughfall to bulk deposition ratio below 1.0 for the N compounds however, also indicates additional input by stemflow and the possible occurrence of canopy uptake. The input of stemflow and the rate of canopy exchange (uptake) in the case of N ( $\text{NH}_4$ ) was thus calculated to derive total deposition (see Section 6.2.4).

Table 6.7 Throughfall to bulk deposition ratios for SO<sub>4</sub>, N, NH<sub>4</sub> and NO<sub>3</sub> at 144 Intensive Monitoring Plots.

Statistic	Throughfall to bulk deposition ratio			
	SO <sub>4</sub>	N	NH <sub>4</sub>	NO <sub>3</sub>
5%	0.72	0.35	0.30	0.31
50%	1.37	0.90	0.81	0.94
95%	2.41	2.52	2.75	2.77
slope <sup>1)</sup>	1.54	1.32	1.21	1.33
intercept <sup>2)</sup>				
R <sup>2</sup> <sub>adj</sub>	75	59	49	61

<sup>1)</sup> Refers to the slope of a linear regression relationship with R<sup>2</sup><sub>adj</sub> being the percentage of variance accounted for.

<sup>2)</sup> Intercepts (mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>) appeared to be negative for N compounds, indicating N uptake, but they were statistically insignificant.

Stemflow has only been measured at 23 plots, whereas annual fluxes were calculated at 19 plots. The ratio of stemflow and throughfall at those plots is given in Table 6.8.

Table 6.8. Ranges of the ratio between annual fluxes of S and N compounds in stemflow and throughfall at 19 Intensive Monitoring plots

Statistic	Stemflow to throughfall ratio			
	SO <sub>4</sub>	NH <sub>4</sub>	NO <sub>3</sub>	N
5%	0.021	0.011	0.002	0.004
50%	0.123	0.068	0.084	0.085
95%	0.438	0.551	0.328	0.458

Ratios mostly ranged between 0.0 and 0.5 with median values near 0.1. Even though the calculated ratios at those plots were in the same order of magnitude, there was no correlation between measured and calculated stemflow with the exception of H, Na and Cl (R<sup>2</sup><sub>adj</sub> = 54%, 42% and 42%, respectively). The slope of the regression line was near 1.0. A similar slope was derived for the other ions with the exception of NH<sub>4</sub> and K, with a slope near 0.5. This result implies that an overall indication of the input by stemflow at all other plots can thus reasonably be derived with Eqs. (6.6) and 6.7). At individual plots, however, the calculated stemflow may deviate strongly from the real stemflow.

A correction for the uptake of NH<sub>4</sub> has been included according to the formulas described in Section 6.2.3 (Eqs. 6.8-6.16). Actually, the formulas of Johnson and Lindberg (1992) used in the calculation procedure refer to total N uptake. The low throughfall/bulk ratios for NO<sub>3</sub> does indicate that the assumption of negligible NO<sub>3</sub> uptake may be questionable. Results of the calculations are therefore presented as the uptake of both NH<sub>4</sub> and NO<sub>3</sub>. Results showed a range in N (NH<sub>4</sub>) uptake varying between 100 and 800 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> (the accepted maximum uptake for spruce/fir forests) with a median value of 400 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> (approximately 5 kg.ha<sup>-1</sup>.yr<sup>-1</sup>; Table 6.9). According to the calculation procedure, higher maximum uptake values were calculated for spruce/fir forests than for pine forests and broadleaves. Estimates were limited to plots, where canopy uptake of nitrogen was calculated assuming that canopy leaching of N never occurs. This was limited to 77 plots where base cation leaching (assumed to be partly counteracted by ammonium uptake) occurs (see the following section on base cations).

Table 6.9 Ranges in calculated canopy uptake fluxes of N at 77 Intensive Monitoring plots.

Type of forest	N uptake flux (mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )				
	min	5%	50%	95%	max
Spruce/fir	51	64	273	758	800
Pine/broadleaves	32	76	400	400	400
All forests	32	68	348	612	800

Unlike throughfall, the ratio of the calculated total deposition compared to bulk deposition appeared to be comparable for the N and S compounds (Fig. 6.10). Ninety percent of the ratios ranged between 0.78 and 2.79 for S with a median value of 1.57 and between 0.62 and 3.21 for N with a median value of 1.58.

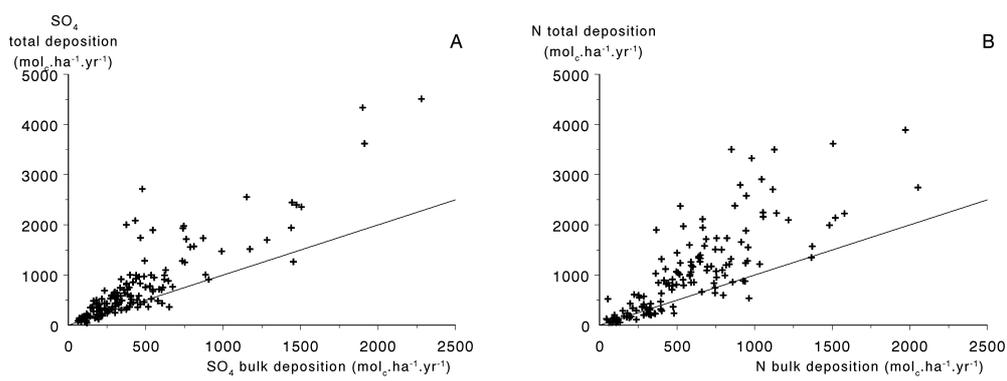


Figure 6.10 Relationship between the calculated total deposition and the measured bulk deposition of SO<sub>4</sub> (A) and N (B). The solid line represents the 1:1 line.

### Base cations

As expected, annual throughfall fluxes of base cations were mostly higher than bulk deposition. However, at a significant number of plots, the input of base cations by throughfall appeared to be lower than by bulk deposition with the exception of K (Fig. 6.11)

As with the S and N compounds, this can be illustrated by (i) the range in throughfall to bulk deposition ratios for the various elements and (ii) the regression coefficients that were derived from a simple linear regression between throughfall and bulk deposition (Table 6.10).

Table 6.10 Throughfall to bulk deposition ratios for Na, Ca, Mg and K at 144 Intensive Monitoring Plots.

Statistic	Throughfall to bulk deposition ratio			
	Na	Ca	Mg	K
5%	0.42	0.79	0.72	1.40
50%	1.34	1.84	1.81	6.51
95%	2.56	4.10	8.38	21.7
slope	1.03	0.92	0.93	1.28
intercept <sup>1)</sup>	80	157	101	285
R <sup>2</sup> <sub>adj</sub> <sup>2)</sup>	65	85	55	9

<sup>1)</sup> Intercepts in mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. Except for Na, intercepts were highly significant.

<sup>2)</sup> R<sup>2</sup><sub>adj</sub> is the percentage of variance accounted for.

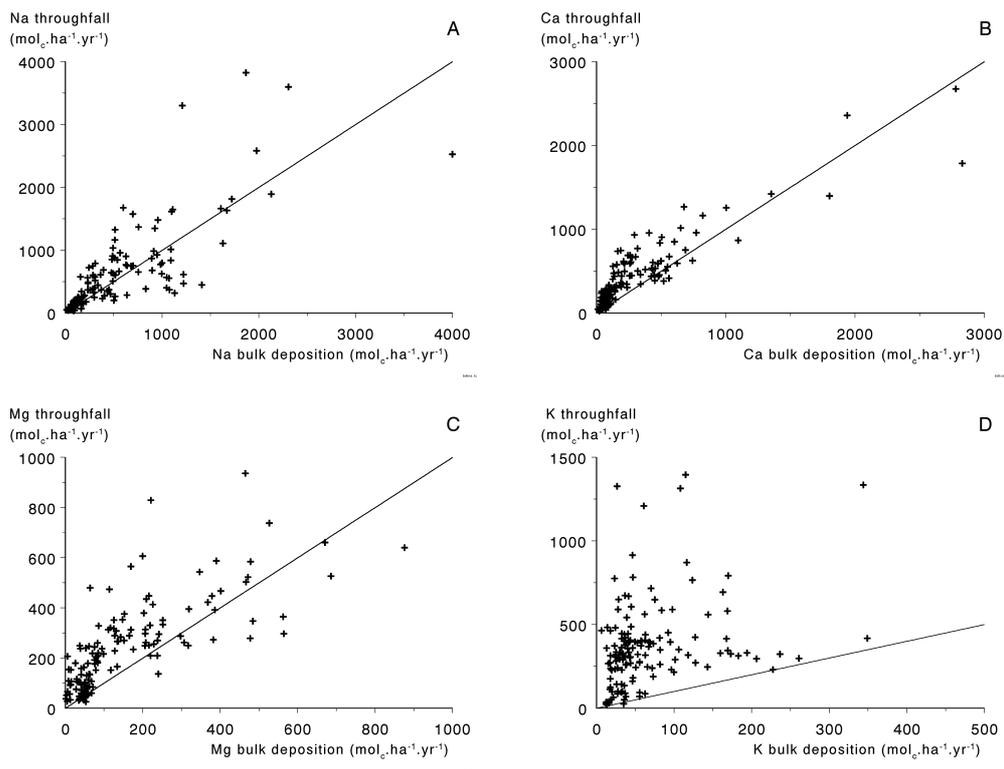


Figure 6.11 Relationship between the annual input by throughfall and bulk deposition for Na (A), Ca (B), Mg (C) and K (D) at 144 Intensive Monitoring plots. The solid line represents the 1:1 line.

For K, there was hardly a relationship between throughfall and bulk deposition ( $R^2_{\text{adj}} = 9\%$ ), indicating that canopy leaching is the dominating process determining the K throughfall flux. A lower input of base cations in throughfall compared to bulk deposition might partly be due to the neglect of stemflow. However, the inputs of base cations in stemflow measured at 19 plots was low. Comparable to the S and N compounds, the ratio of stemflow to throughfall was mostly less than 0.4, with median values near 0.1.

Even the ratio of base cations in both (measured) throughfall and (calculated) stemflow compared to bulk deposition was less than 1.0 at several plots, specifically for Na. This was assumed to be due to measuring inadequacies, because it is unlikely that Na does interact with the forest canopy (Bredemeier, 1998). In calculating canopy exchange, and thereby total deposition of base cations, plots in which the ratio of Na in throughfall and stemflow compared to bulk deposition was lower than 1.0 were therefore neglected. This led to a reduction of almost 25% of the 144 plots. Ranges in cation exchange fluxes thus calculated are given in Table 6.11.

Table 6.11 Ranges in calculated canopy leaching fluxes of base cations at 102 Intensive Monitoring plots.

Statistic	canopy leaching flux ( $\text{mol}_c.\text{ha}^{-1}.\text{yr}^{-1}$ )			
	Ca	Mg	K	BC <sup>1)</sup>
5%	-84	-29	10	-15
50%	60	44	297	391
95%	373	231	828	1272

<sup>1)</sup> Sum of Ca, Mg and K

Negative canopy leaching fluxes were also observed for Ca and Mg, indicating the occurrence of canopy uptake. Irreversible uptake of Ca and Mg, has been reported in the literature (Abrahamsen et al., 1976; Alcock and Morton, 1981). Differences in magnitude and direction (uptake or

leaching) of the base cation canopy exchange is probably dependent on the nutritional status of the trees, absorbing cations in case of base cation deficiencies.

Ratios between total deposition and bulk deposition of Ca, Mg and K were comparable to the ratios between Na in throughfall and in bulk deposition (see Fig. 6.11A and Table 6.10), since those ratios were the basis for the calculation of total base cation deposition (see Eq. 6.8). However, by neglecting the plots where Na in throughfall (and stemflow) was less than in bulk deposition, the ratios increased substantially. 90% of the values ranged between 0.46 and 2.75, with a median value of 1.47.

### 6.3.2 Ion ratios in bulk deposition, throughfall and total deposition

#### *Sulphur versus nitrogen compounds*

Results showed a significant correlation between the input of N and S at the various Intensive Monitoring plots, especially for the 19 plots where stemflow was measured (Fig 6.12).

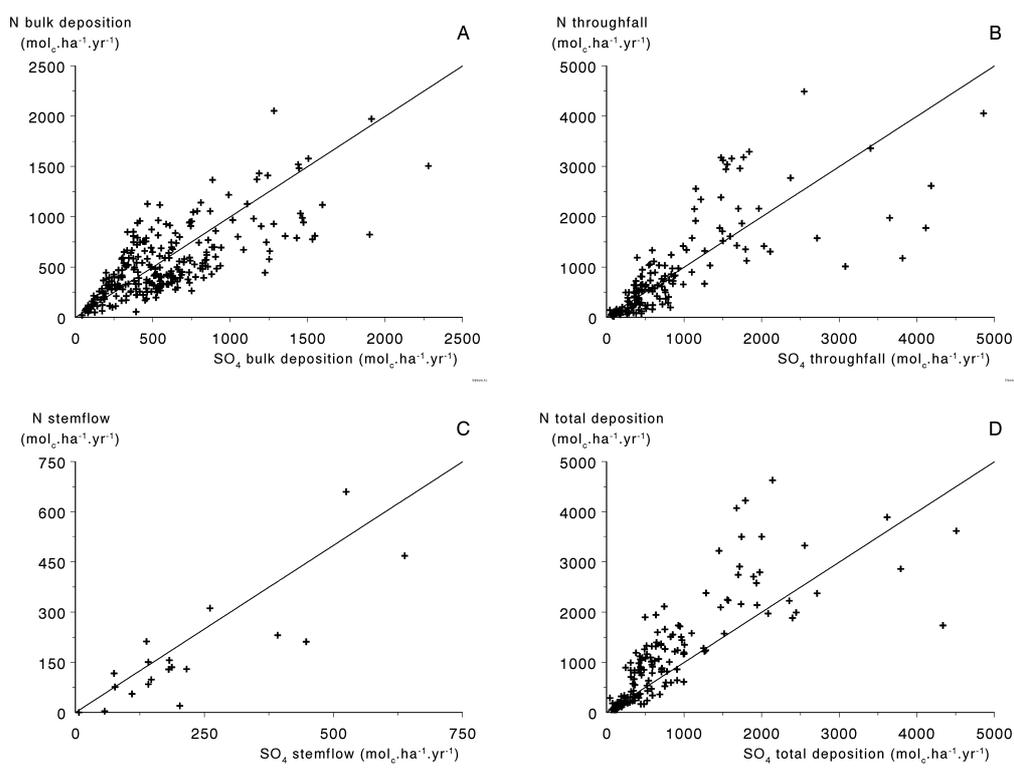


Figure 6.12 Relationships between the annual fluxes of N and S in bulk deposition plots (A; 268 plots), throughfall (B; 163 plots), stemflow (C; 19 plots) and total deposition (D; 144 plots). The solid line represents the 1:1 line.

The percentage variance accounted for in linear regression relationships equalled 52% for bulk deposition, 51% for throughfall, 69% for stemflow and 63% in total deposition. On average, the N input in bulk deposition and throughfall equalled the S deposition, but the N to S ratios ranged from approximately 0.3 to 2.0 (Table 6.12). Assuming that the calculated N uptake rates are reasonable, the average N to S ratio in total deposition is, however, higher than 1.0. This result is

influenced by the different number of plots for total deposition (144) compared to throughfall (163), but this influence appeared to be very small.

Table 6.12 Ranges in the N to S ratio in bulk deposition, throughfall and total deposition at the Intensive Monitoring plots.

Statistic	N/S ratio		
	Bulk deposition (N = 268)	Throughfall (N = 163)	Total deposition (N = 144)
mean	1.03	1.02	1.46
5%	0.46	0.27	0.52
50%	0.95	0.93	1.28
95%	1.97	1.97	2.70

The comparatively high N deposition and the significant relationship with S deposition at a European wide scale is a striking result. In the eighties, S emissions were generally considered the most important cause of acid deposition. However, since then S emissions and thereby S deposition have constantly decreased over large parts of Europe whereas N emissions mostly stayed constant or even increased. This causes N to be a dominating factor in the acidic input in large parts of Europe (see also Section 6.3.3). The relationship between N and S deposition points toward co-emission from SO<sub>x</sub> (mainly industry), NO<sub>x</sub> (mainly traffic) and NH<sub>x</sub> (mainly agriculture) in industrialised areas. Furthermore, the correlation between the input of NH<sub>4</sub> and SO<sub>4</sub> (not shown) may be influenced by co-precipitation of both ions (e.g. Van Breemen et al., 1982).

The relative contribution of NH<sub>4</sub> and NO<sub>3</sub> in N deposition varies largely over the plots (Table 6.13) but in general there is a weak but significant correlation between both N compounds (Fig. 6.13).

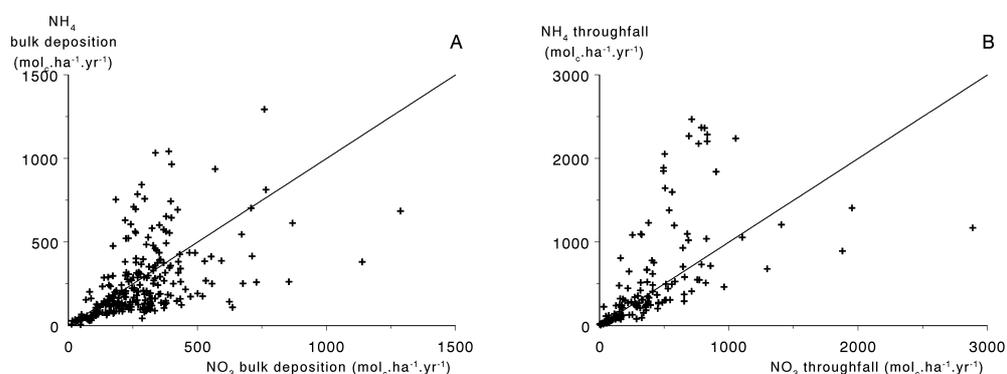


Figure 6.13 Relationships between the annual fluxes of NH<sub>4</sub> and NO<sub>3</sub> in bulk deposition (A; 286 plots) and throughfall (B; 163 plots). The solid line represents the 1:1 line.

On average, the NH<sub>4</sub> and NO<sub>3</sub> fluxes in bulk deposition were about equal at the 268 plots where bulk deposition is measured (ratio is 0.95). This is significantly lower than the NH<sub>4</sub> to NO<sub>3</sub> ratio of 1.36 in throughfall. This difference is, however, due to the 122 bulk deposition plots in Poland, with an average NH<sub>4</sub> to NO<sub>3</sub> ratio of 0.53. The NH<sub>4</sub> to NO<sub>3</sub> ratio at the remaining 146 plots equals 1.30. This is comparable to the mean NH<sub>4</sub> to NO<sub>3</sub> ratio in throughfall at those plots (Table 6.13). The NH<sub>4</sub> to NO<sub>3</sub> ratio in total deposition is likely to be even higher, but this ratio has not been given considering the uncertainties in the assumption that all N is taken up as NH<sub>4</sub> (see before).

Table 6.13 Ranges in the  $\text{NH}_4$  to  $\text{NO}_3$  ratio in bulk deposition, throughfall and stemflow at the Intensive Monitoring plots.

Statistic	$\text{NH}_4/\text{NO}_3$ ratio		
	Bulk deposition (N = 268)	Throughfall (N = 163)	Stemflow (N = 19)
mean	0.95	1.36	1.75
5%	0.33	0.48	0.43
50%	0.81	0.91	0.94
95%	2.34	3.42	6.92

The percentage of variance accounted for in the relationships between  $\text{NH}_4$  and  $\text{NO}_3$  equals 27% for bulk deposition (correlation coefficient is 0.56) and 37% for throughfall (correlation coefficient is 0.65).

### Base cations versus acidic compounds

Whether the input of S and N compounds from the atmosphere causes acidification is strongly influenced by the deposition of accompanying base cations. The relationship between Cl corrected base cation deposition ( $\text{Ca} + \text{Mg} + \text{K} + \text{Na} - \text{Cl}$ ) and the sum of N and S deposition at the Intensive Monitoring plots is quite low, especially for bulk deposition (Fig 6.14). In bulk deposition, the input of base cations (corrected for Cl) is often higher than the S and N input (Fig. 6.14A) but in the total deposition, this is generally not the case (Fig 6.14B). This is due to the larger input of dry deposition of S and N compounds compared to base cations.

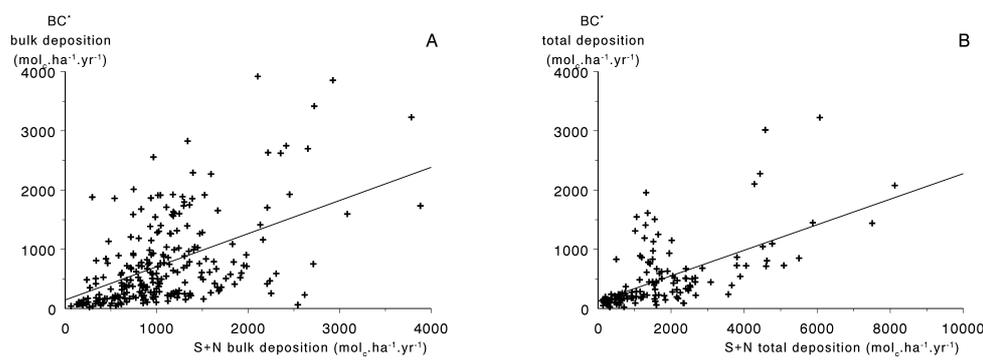


Figure 6.14 Relationships between the annual fluxes of chloride corrected base cations and S+N in bulk deposition (A; 286 plots) and total deposition (B; 144 plots). The solid line represents a regression line.

A more significant relationship was observed between the deposition of Ca and  $\text{SO}_4$  in total deposition (Fig. 6.15). The percentage variance accounted for in linear regression relationships was 40% for bulk deposition (correlative coefficient is 0.67) and 57% for total deposition (correlative coefficient is 0.78). The correlation may partly be due to associated emissions of  $\text{SO}_2$  and Ca from smelters and refineries, whereas recent investigations also indicate co-precipitation of Ca and  $\text{SO}_4$ .

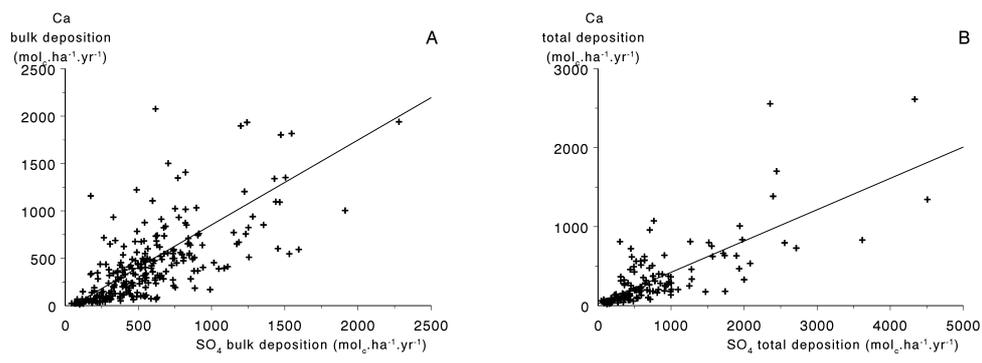


Figure 6.15 Relationships between the annual fluxes of Ca and SO<sub>4</sub> in bulk deposition (A; 286 plots) and total deposition (B; 144 plots). The solid line represents a regression line.

An overview of the ranges in the ratios between base cation inputs and S and N inputs (Table 6.14) further illustrates that the impact of base cations on the potential acid input can be very large at certain plots. It sometimes can lead to a negative acid input, since the base cation deposition is larger than the sum of S and N deposition. In total deposition, however, this occurs only at 5 – 10% of the plots.

Table 6.14 Ranges in base cation to S+N ratios in bulk deposition and total deposition at the Intensive Monitoring plots.

Statistic	BC <sup>+</sup> to S+N ratio		Ca/SO <sub>4</sub> ratio	
	Bulk deposition (N = 268)	Total deposition (N = 144)	Bulk deposition (N = 268)	Total deposition (N = 144)
mean	0.74	0.36	0.78	0.47
5%	0.12	0.04	0.17	0.14
50%	0.51	0.26	0.60	0.37
95%	1.99	1.04	1.89	1.11

### 6.3.3 Ranges and geographic variation in bulk deposition, throughfall and total deposition

#### *Bulk deposition*

Bulk deposition of both S and N varied mostly (approximately 90% of the values) between 100 and 1400 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. The bulk deposition of acidity, being the sum of both compounds, corrected for the input of base cations, ranged from less than 0 (the input of base cations is higher than the deposition of S and N) to more than 2000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> (Fig. 6.16A). A similar range was observed for the sum of the base cations corrected for chloride (Ca+Mg+K+Na-Cl) that neutralises the potential acid input (Fig. 6.16B).

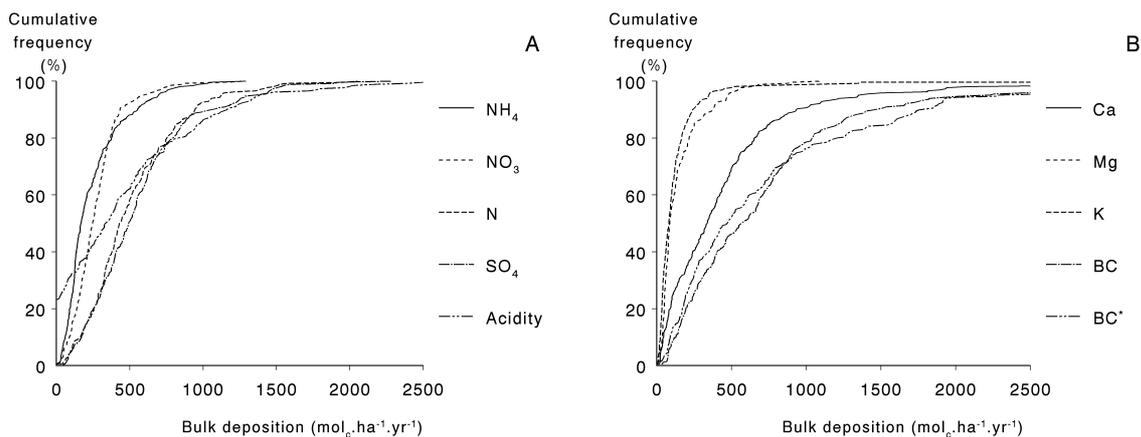


Figure 6.16 Cumulative frequency distributions of the bulk deposition of S, N and acidity (A) and of base cations (B) at 268 Intensive Monitoring plots.

In comparison to throughfall, the range in bulk deposition in Fig. 6.16 is influenced by the occurrence of 122 bulk deposition plots in Poland. Data on the ranges in bulk deposition in Poland and at the remaining Intensive Monitoring plots show, however, a comparable range for most elements in both areas (Fig. 6.17).

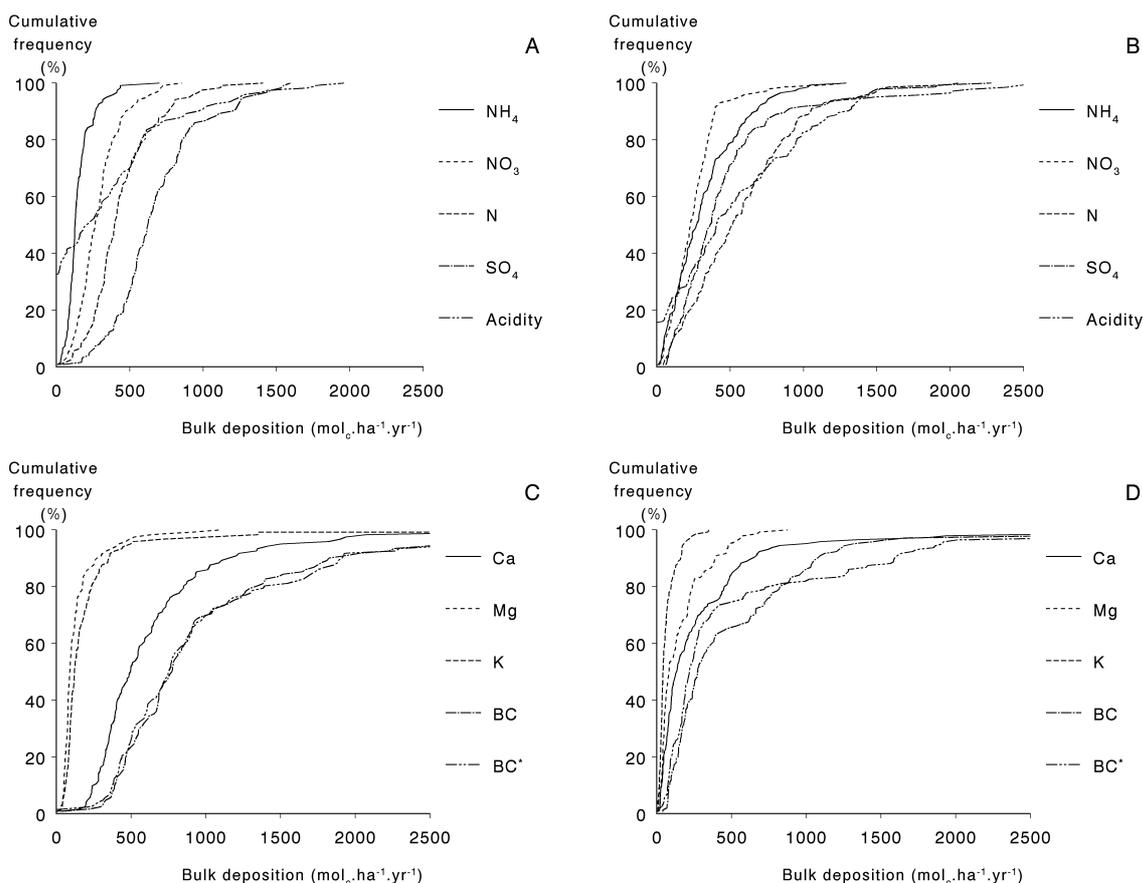


Figure 6.17 Cumulative frequency distributions of the bulk deposition of S, N, and acidity in Poland (A; 122 plots) and in the remaining countries (B; 146 plots) and of base cations in Poland (C; 122 plots) and in the remaining countries (D; 146 plots).

Even in a high S deposition area, such as Poland, there appears to be a wide range in S deposition from less than 200 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> (approximately 3 kg.ha<sup>-1</sup>.yr<sup>-1</sup>) to more than 1000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> (16 kg.ha<sup>-1</sup>.yr<sup>-1</sup>). A comparable range was also observed for N compounds illustrating the relationship between N and S deposition, discussed in Section 6.3.2. The influence of geographic region on the annual average bulk deposition fluxes is illustrated in Table 6.15.

*Table 6.15 Annual average bulk deposition fluxes of major elements as a function of geographic region*

Region	N <sup>1)</sup>	Bulk deposition flux (mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )						
		SO <sub>4</sub>	NO <sub>3</sub>	NH <sub>4</sub>	Ca	Mg	K	Na
North/Boreal	40	170	106	95	49	54	32	154
North/Boreal temperate	11	239	184	153	71	41	31	136
West/Atlantic	47	500	280	435	191	183	57	778
Central/East	143	703	311	197	666	150	196	186
South/Mediterranean	27	628	334	442	618	285	130	868

<sup>1)</sup> N = number of plots; total = 268.

In general, the deposition of all compounds is lowest in Northern Europe, even though there is a clear gradient in the deposition of S and N compounds and of Ca, going from Northern Scandinavia (Boreal) to Southern Scandinavia (Boreal Temperate, being Southern Sweden and Southernmost Norway). As expected, deposition of S and N compounds is much higher in Western, Central and Eastern Europe, but it also appears to be high in the Mediterranean area. This result is most likely biased by the uneven distribution of plots (see also Figs. 6.18 and 6.19). Furthermore, results for throughfall (see Table 6.16) indicate that dry deposition plays a much larger role in the deposition of S and N compounds in Western and Central Europe than in the Mediterranean area. The deposition of Ca and K is highest in Central/Eastern Europe and in the Mediterranean area, whereas Na is most important in countries located near the sea (Western Europe and the Mediterranean area).

The influence of geographic region is also illustrated on maps of the bulk deposition of SO<sub>4</sub> (Fig. 6.18) and N (Fig. 6.19) at the Intensive monitoring plots. The maps show low deposition values (<200 – 400 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>) of both S and N at plots in Northern Europe, with the exception of the southern part of Norway and Sweden where the bulk deposition of both compounds mostly ranges between 400 and 800 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. Bulk deposition of N generally appeared to be higher than S deposition at plots in Western Europe (UK, Belgium, Netherlands, Luxembourg, France), whereas the reverse was generally observed at plots in Central Europe (Poland, Czech Republic, Austria, Hungary; Compare Fig 6.18 and 6.19). In general, there was, however, a clear correlation between N and S deposition, as indicated before (Section 6.3.2).

The geographic pattern of bulk deposition of Ca, being the most important base cation neutralising the potential acid input from the atmosphere is quite comparable to S and N, being low in Northern Europe, high in Central and Southern Europe and Intermediate in Western Europe (Fig. 6.20). This is consistent with the correlation between Ca and SO<sub>4</sub> deposition mentioned before (Section 6.3.2).

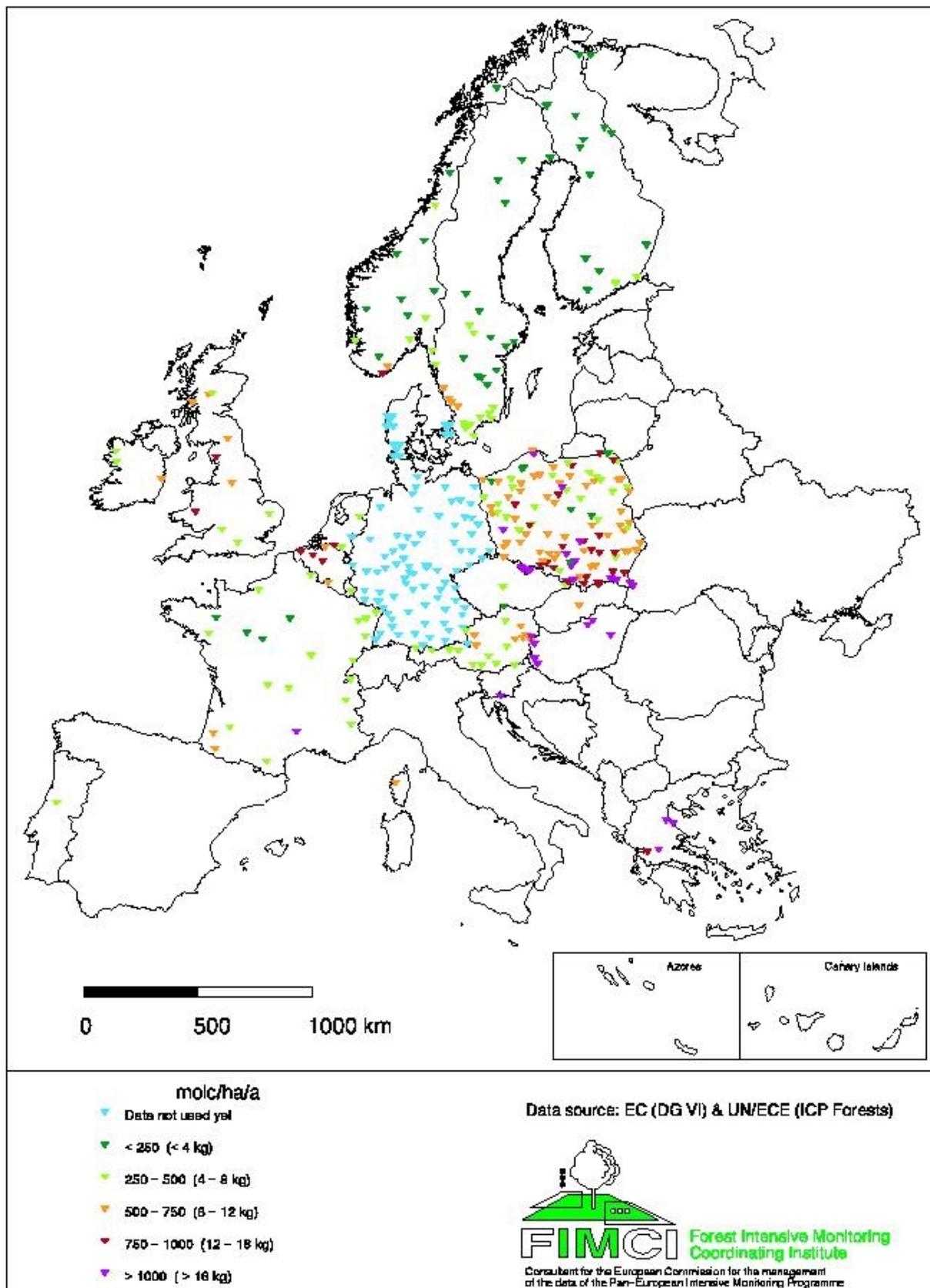


Figure 6.18 Geographical variation of the bulk deposition of S in 1996 at 268 Intensive Monitoring plots

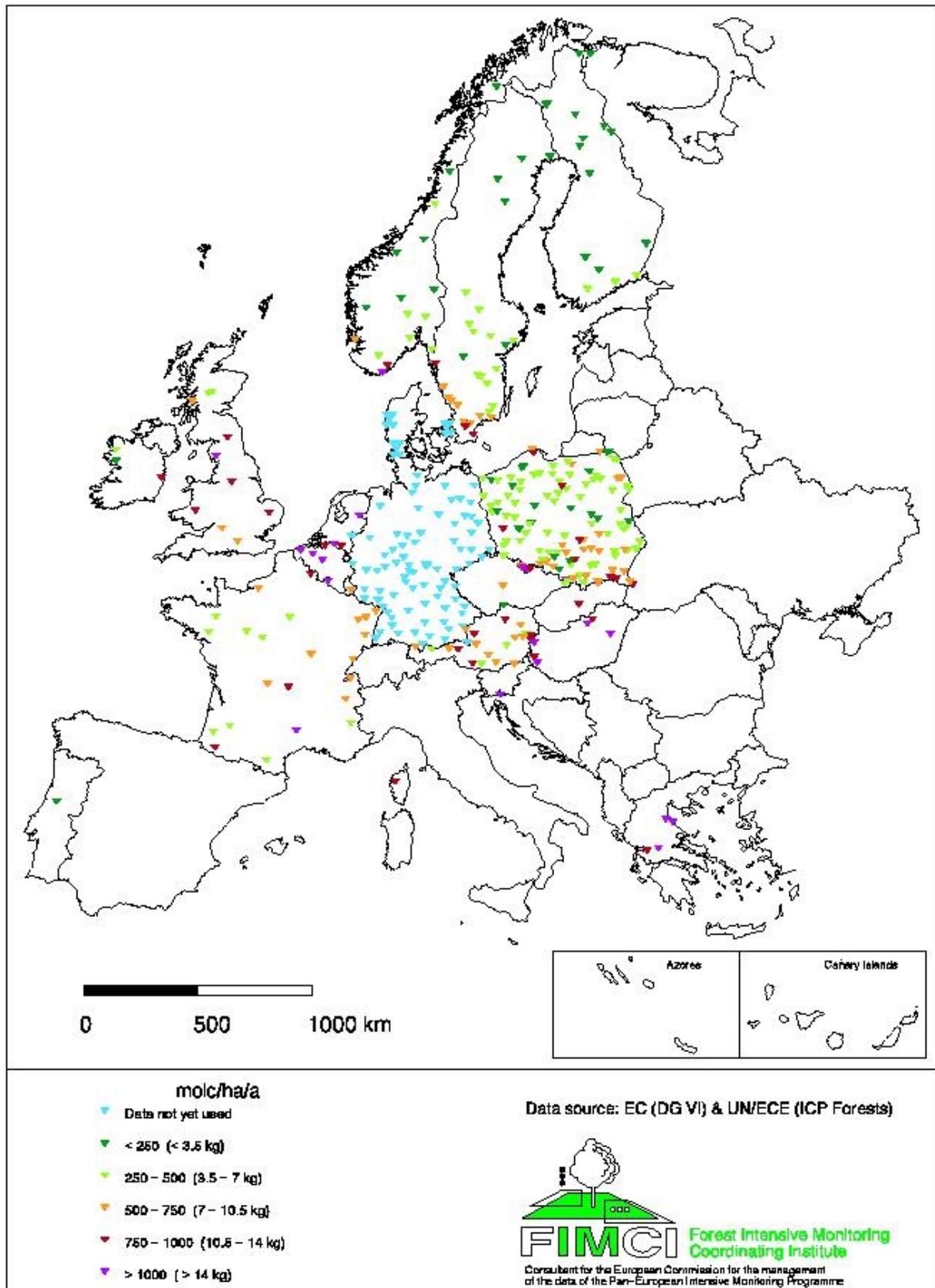


Figure 6.19 Geographical variation of the bulk deposition of N in 1996 at 268 Intensive Monitoring plots

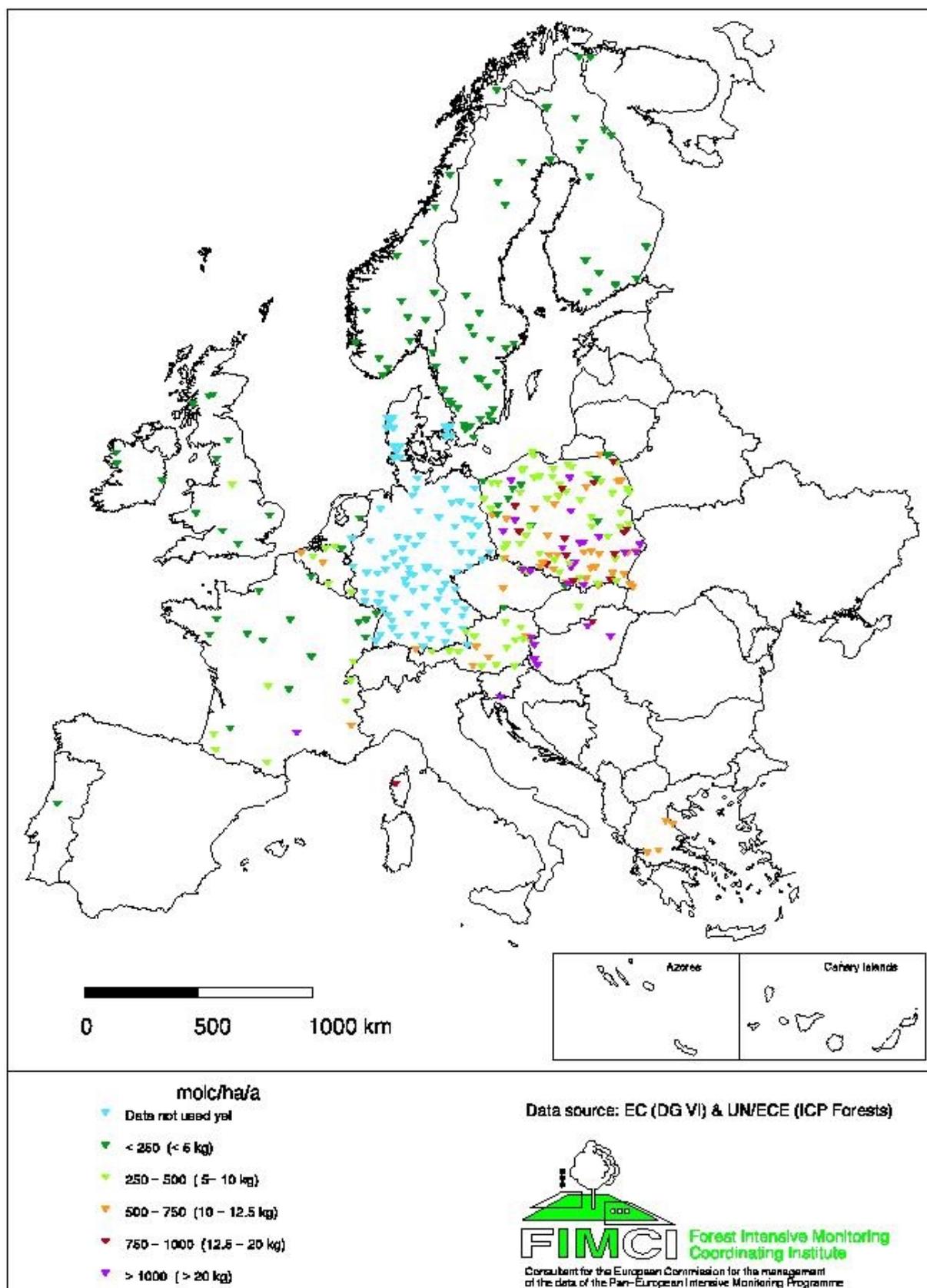


Figure 6.20 Geographical variation of the bulk deposition of Ca in 1996 at 268 Intensive Monitoring plots

## Throughfall

Throughfall fluxes of N and S were quite comparable to bulk deposition. Values mostly ranged between 100 and 3000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> but throughfall fluxes up to 4000-8000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> were also observed for N and S, respectively (Fig. 6.21).

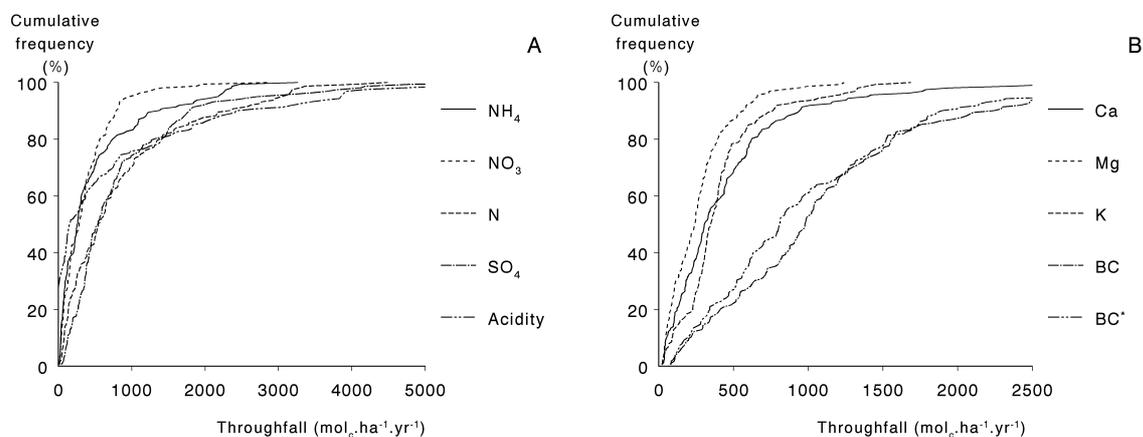


Figure 6.21 Cumulative frequency distributions of the throughfall of S, N and acidity (A) and of base cations (B) at 163 Intensive Monitoring plots.

The geographic variation of throughfall fluxes of S (Fig. 6.22) and N compounds (Fig. 6.23) is quite comparable to bulk deposition at similar plots (Compare Fig. 6.18 and Fig. 6.19). The impact of the difference in plots is illustrated by comparing Table 6.15 and 6.16. The throughfall fluxes for SO<sub>4</sub> and NO<sub>3</sub> are now highest in Central/Eastern Europe, whereas NH<sub>4</sub> deposition is highest in Western Europe. This pattern seems most consistent with the expectations. It illustrates that conclusions about the influence of a region has to be considered with care, due to the uneven representation of plots in the various regions.

Table 6.16 Annual average throughfall fluxes of major elements as a function of geographic region

Region	N <sup>1)</sup>	Throughfall flux (mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )						
		SO <sub>4</sub>	NO <sub>3</sub>	NH <sub>4</sub>	Ca	Mg	K	Na
North/Boreal	40	235	78	64	103	96	154	263
North/Boreal temperate	11	344	116	88	157	112	269	230
West/Atlantic	60	981	428	887	405	358	511	1037
Central/East	26	2068	819	640	989	365	618	303
South/Mediterranean	26	657	355	307	756	321	429	707

<sup>1)</sup> N = number of plots; total = 163.

Results show high inputs of both S and N at plots in the Netherlands, Belgium and Luxembourg but also in the Czech Republic, Slovak Republic and Hungary. This again illustrates that high deposition of S is generally associated with an increased N deposition. In Central Europe the acidic input is largely set off by the input of base cations. (See Fig. 6.20).

## Total deposition

Ranges in total deposition of S and N compounds are comparable to those in throughfall for both compounds (100-3000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> at approximate 90% of the plots). Due to the correction for canopy uptake, the median value for N input increased from 546 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> for throughfall to 889 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> for total deposition (from approximately 8-12.5 kg.ha<sup>-1</sup>.yr<sup>-1</sup>). The total input of acidity ranged mostly between 200-4000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> with extremes up to 10,000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>.

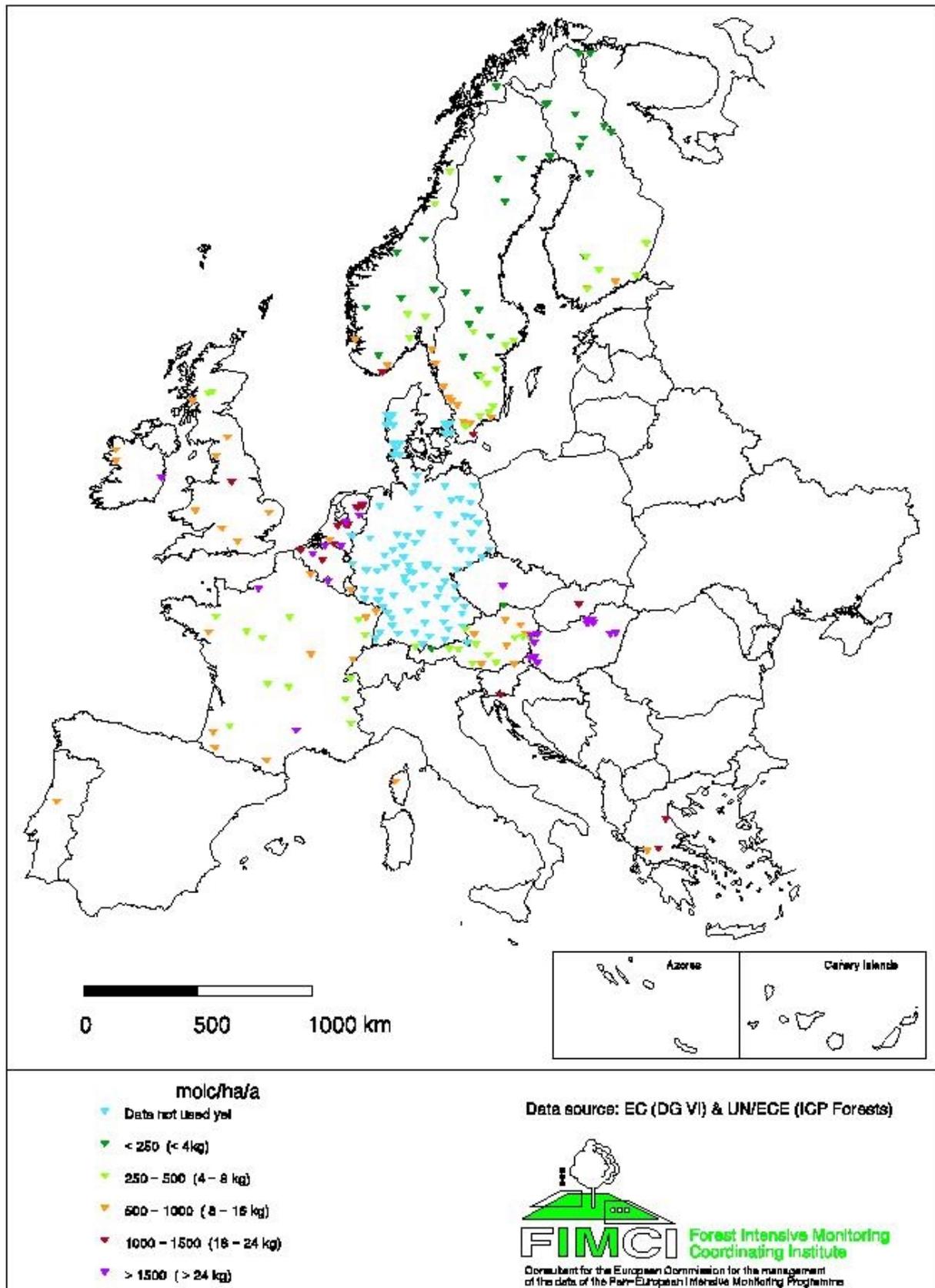


Figure 6.22 Geographical variation of the throughfall of S in 1996 at 163 Intensive Monitoring plots

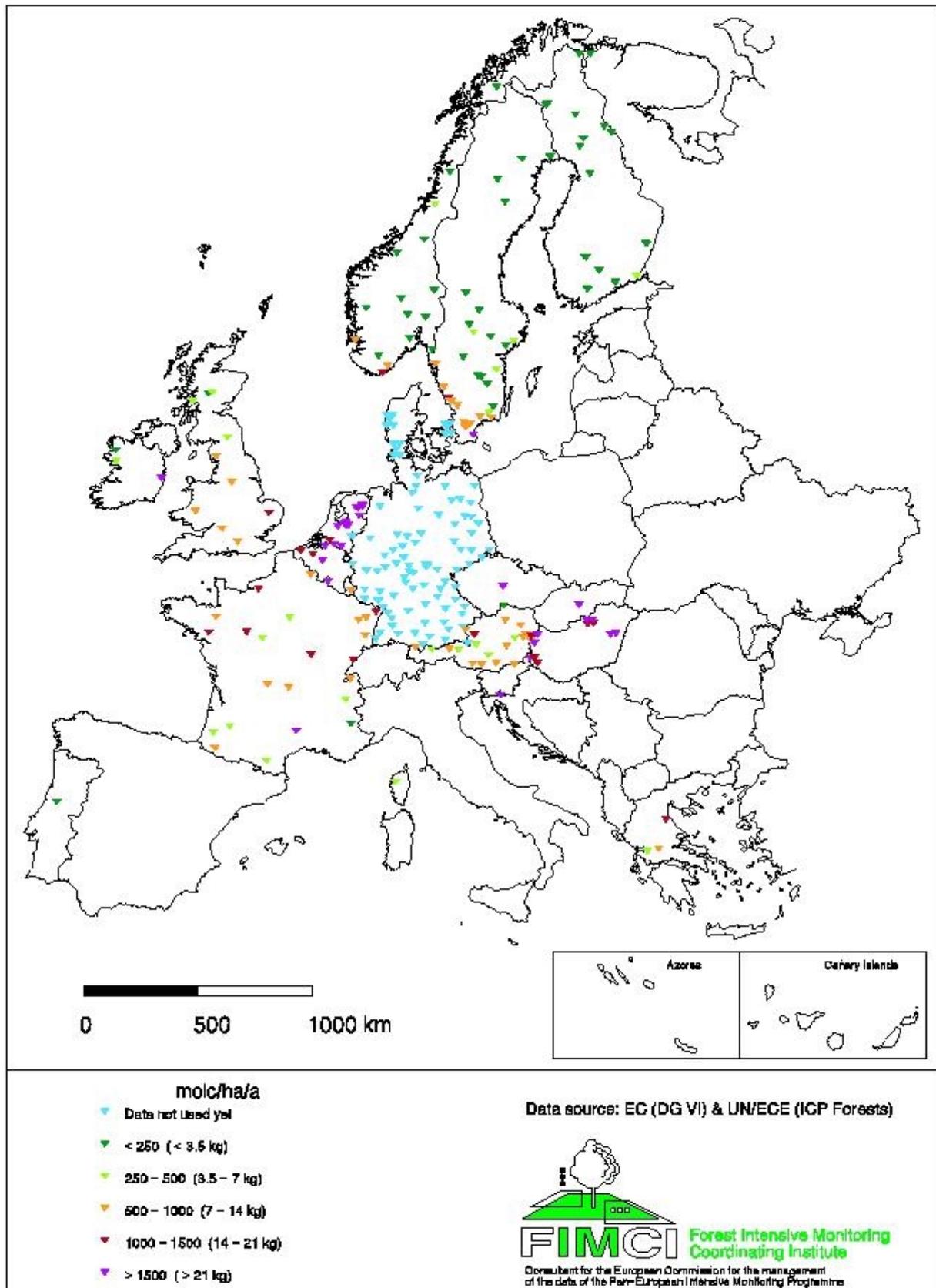


Figure 6.23 Geographical variation of the throughfall of N in 1996 at 163 intensive monitoring plots

In general the input of S and N by stemflow is only 10% of the input by throughfall but N uptake has a significant influence on the calculated N deposition (Section 6.2.1; Fig. 6.24A). For the base cations, the range in total deposition values were more comparable to bulk deposition than to throughfall (mostly between 100 and 2000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> for the sum of Ca, Mg and K) indicating the less significant contribution of dry deposition compared to S and N compounds (Fig. 6.24B).

The impact of geographic location on throughfall fluxes is quite comparable to bulk deposition, despite the large difference in Central/Eastern Europe and Southern Europe (Compare Table 6.15 and 6.17).

Table 6.17 Annual average total deposition fluxes of major elements as a function of geographic region

Region	N <sup>1)</sup>	Total deposition flux (mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )						
		SO <sub>4</sub>	NO <sub>3</sub>	NH <sub>4</sub>	Ca	Mg	K	Na
North/Boreal	33	273	88	148	79	102	52	330
North/Boreal temperate	10	382	128	277	137	86	60	257
West/Atlantic	40	954	419	1027	341	285	104	117
Central/East	10	960	641	1027	435	106	78	329
South/Mediterranean	9	958	444	610	943	339	118	1100

<sup>1)</sup> N = number of plots; total = 102.

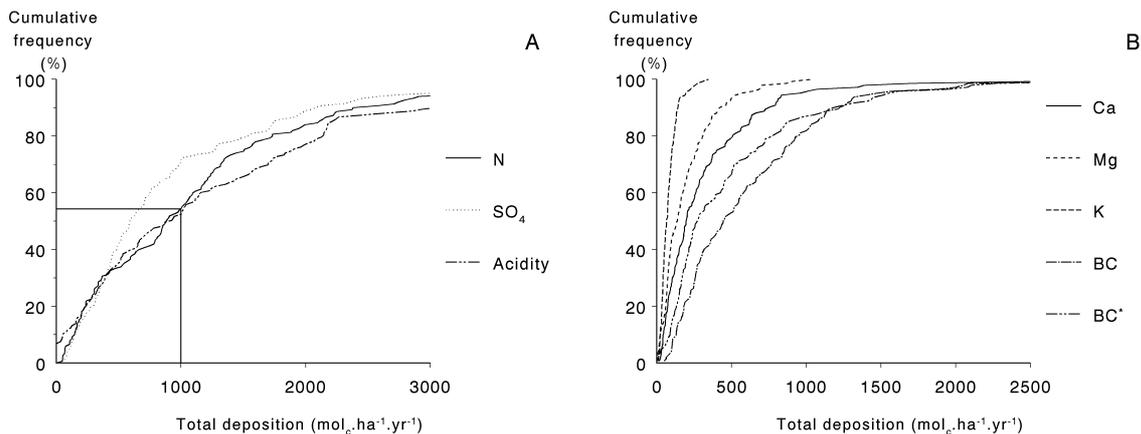


Figure 6.24 Cumulative frequency distributions of the total deposition of S, N and acidity (A) and of base cations (B) at 144 Intensive Monitoring plots.

Critical N loads related to impacts on the species diversity of the ground vegetation of forests generally vary between 1000 and 1500 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> (approximately 15-20 kg.ha<sup>-1</sup>.yr<sup>-1</sup>; Bobbink et al., 1996). Approximately 45% of the considered plots get an external N input above 1000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>, thus being in a state of risk with respect to impacts on ground vegetation (Fig. 6.24A). Critical loads related to impacts on tree health, such as nutrient imbalances and increased shoot-root ratios causing drought stress, vary mostly between 1500 and 3000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>, depending amongst others on the relative contribution of NH<sub>4</sub> and NO<sub>3</sub> deposition, the nitrification rate and buffer rate of the soil and the tree species (De Vries, 1993; Bobbink et al., 1996). Present N loads do occur in that range, thus indicating the possibility of those adverse effects. On the other hand, one has to be aware that N loads below e.g. 1000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> may inhibit tree growth due to N limitation.

Critical acid loads, related to an increased ratio of Al to the base cations Ca, Mg and K vary mostly between 1500 and 3500 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>, depending mainly on the buffer rate of the soil and the sensitivity of the tree species to elevated Al/(Ca+Mg+K) ratios (De Vries, 1996). A more specific comparison of present loads and critical loads is needed to get insight in the possible exceedance of critical acid loads since total atmospheric deposition on most of the plots varied between this range. Data on the present Al/(Ca+Mg+K) ratios in soil solution at part of those plots do, however, indicate that the acid input causes exceedances of the critical Al/(Ca+Mg+K) ratio in 11 - 17% of the cases, depending on the layer considered.

### 6.3.4 Relationships between atmospheric deposition and stand/site characteristics

#### 6.3.4.1 Principal Component Analyses

##### *Relationships with site characteristics for a data set with 125 sites*

Figure 6.25 shows the biplot resulting from the PCA analysis on the deposition data set in combination with major site characteristics (deposition region, tree species and altitude) and precipitation (a total of 125 sites). The first and second axis explain 62 and 15% of the total variance, respectively. The site and stand characteristics explain 44% of the variation in deposition between sites, of which 83% is displayed in the diagram.

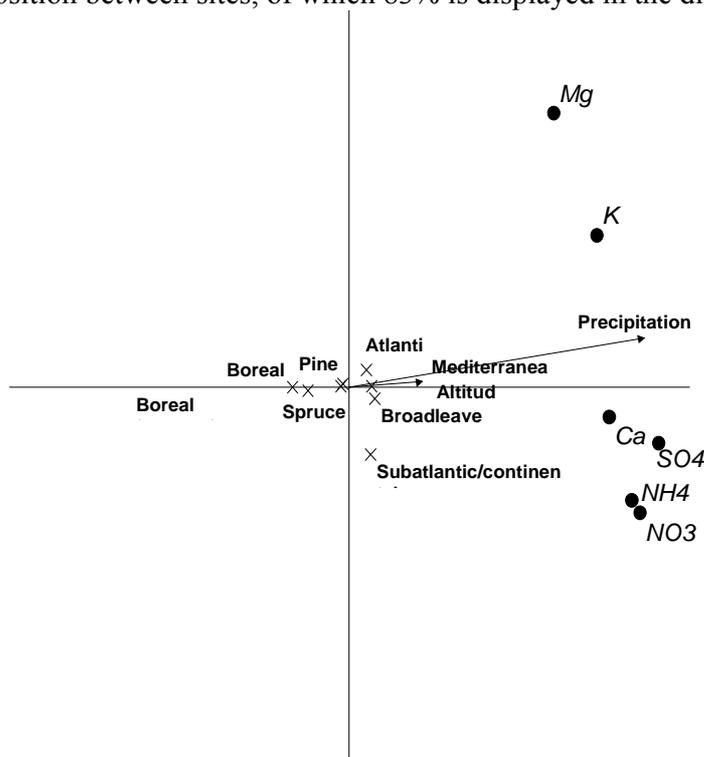


Figure 6.25 PCA diagram of the deposition data (response variables) and different site characteristics and precipitation (predictor variables) for 125 sites. See also Section 3.3.2 for interpretation of the diagram.

The diagram shows that all deposition variables are correlated with each other, especially SO<sub>4</sub>, NH<sub>4</sub> and NO<sub>3</sub> and Ca. As stated before, this correlation between deposition variables indicates the occurrence of co-emission (specifically with respect to SO<sub>4</sub>, NO<sub>3</sub> and Ca) and probably also to co-precipitation in the case of SO<sub>4</sub> and NH<sub>4</sub>. The strong positive correlation of the deposition

variables with precipitation indicates the increase in wet deposition with an increase in rainfall. As expected, the ordination diagram also shows that the geographic region is correlated with deposition. Results indicate that atmospheric deposition is higher in the Atlantic, Subatlantic/Continental and even the Mediterranean regions compared to the Boreal regions. Unlike the expectations, the results seem to indicate that altitude is positively correlated with deposition position. This is, however, due to the correlation between altitude and region. In a multiple regression analyses, where altitude is considered, while accounting for differences in region, the opposite effect is observed (see Section 6.3.4.2). The same effect occurs for tree species. The slightly positive correlation with deciduous tree species is due to their geographic occurrence and not to their filtering effect, which is known to be higher for coniferous trees (see Section 6.3.4.2).

**Relationships with site and stand characteristics for a data set with 35 sites**

Figure 6.26 shows the biplot resulting from the PCA analysis on a deposition data set in combination with both major site characteristics (deposition region, tree species and altitude), precipitation and major stand characteristics (stand height and skewness ; a total of 35 sites only). The first and second axis explain 77 and 15% of the total variance respectively. The site and stand characteristics explain 83% of the variation in deposition between sites, of which 95% is displayed in the diagram.

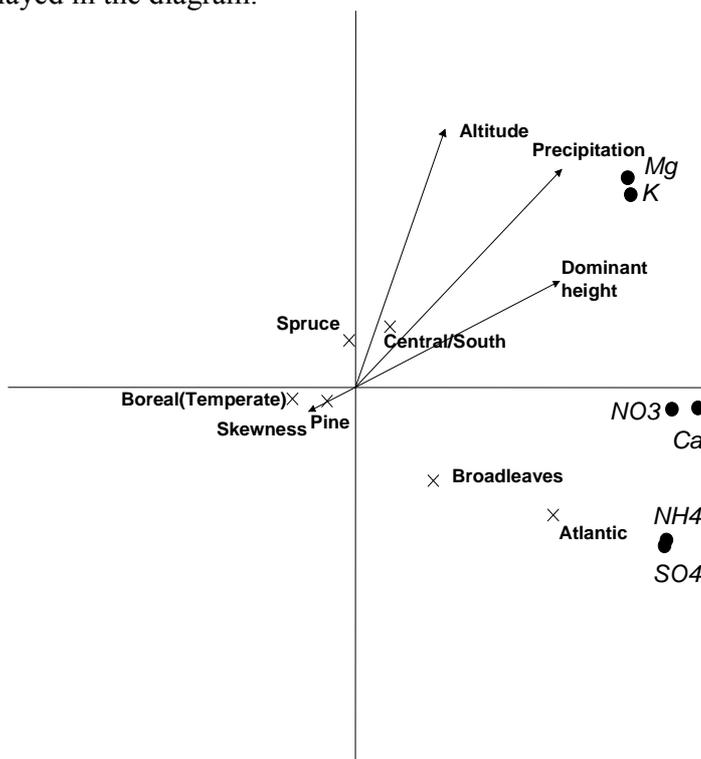


Figure 6.26 PCA diagram of the deposition data (response variables) and different site and stand characteristics and precipitation (predictor variables) for 35 sites. See also Section 3.3.2 for interpretation of the diagram.

The diagram shows the same correlations among deposition parameters as indicated by the former PCA analysis. Also their relation with precipitation and region is unchanged. The positive correlation relation between deposition and dominant height can be explained from an increasing dry deposition with an increasing dominant height of the trees. Strange enough, this result was not found in the regression analyses. No strong correlation between deposition and skewness is indicated.

### 6.3.4.2 Regression analyses

Results of a multiple regression analyses of the deposition data on the various environmental factors is shown in Table 6.18. The regression analyses included both qualitative variables (geographic region and tree species) and quantitative variables (altitude and precipitation). The results are related to the dataset in which stand characteristics (tree height, stand density index etc) have not been included.

Table 6.18 Overview of the predictor variables explaining the deposition of  $SO_4$ ,  $NO_3$ ,  $NH_4$ , N, Ca, Mg, K and BC (sum of Ca, Mg and K).

Predictor variables	$SO_4$	$NO_3$	$NH_4$	N	Ca	Mg	K	BC
<i>Region</i>								
West Atlantic <sup>2)</sup>	--	--	--	--	--	--	--	--
North/Boreal	--	--	--	--	0	--	-	--
North/Boreal temperate	--	--	--	--	0	--	-	--
Central/East	++	++	0	+	+	-	0	0
South/Mediterranean	0	0	0	0	0	0	0	0
Precipitation	+	++	0	+	++	++	0	++
<i>Site/stand</i>								
Altitude	--	--	--	--	0	-	0	0
Spruce	0	0	0	0	-	0	0	0
Broadleaves	0	-	0	-	+	0	0	0
N <sup>2)</sup>	160	160	160	160	122	122	122	122
R <sup>2</sup> <sub>adj</sub> (%) <sup>3)</sup>	37	35	42	39	60	37	20	59

<sup>1)</sup> 0 = insignificant at the 95% level: t value < 2.0

+/- = significant: t value > 2.0

++/-- = highly significant: t value > 3.0

A '+' sign implies that the response variable (the concentration of  $SO_4$ ,  $NO_3$ ,  $NH_4$  or total N) increases with an increase in the predictor variable, whereas a '-' sign implies the opposite. Signs in brackets are related to results in which the logarithmic concentrations were used against the logarithm of the deposition and precipitation (excess).

<sup>2)</sup> Reference is pine forest in the Atlantic region

<sup>3)</sup> N = number of plots.

<sup>4)</sup> R<sup>2</sup><sub>adj</sub> = percentage variance accounted for.

The results for the geographic region are related to Western Europe (Atlantic region), while the effect of the tree species is related to pine forests. As with the results of the PCA, the regression results indicate that atmospheric deposition of all ions is significantly lower in the Boreal regions compared to Western Europe (the Atlantic region), with the exception of Ca deposition in southern Sweden and southernmost Norway (Boreal temperate). This holds also for Central/Eastern Europe (Subatlantic/Continental region) and Southern Europe (the Mediterranean region). Another clear result is that  $SO_4$ ,  $NO_3$  and Ca deposition is significantly higher in the Central/Eastern part of Europe compared to Western Europe.

The strong positive correlation of most deposition variables with precipitation indicates the increase in wet deposition with an increase in rainfall. For  $NH_4$  and K, the relationship is however not significant. Unlike the PCA, the results now indicate that altitude is significantly negatively correlated with the deposition of S and N compounds, since the regression analyses accounts for differences in region. The impact of altitude on base cation deposition is, however, insignificant. Impacts of tree species on the element deposition is mostly insignificant in this dataset, since geographic region overwhelms the variation. The positive influence of deciduous trees on Ca deposition is likely to be an artefact due to the correlation between tree species and region. In a

multiple regression analyses using the ratios of total deposition to bulk deposition as the response variable, indicative for the occurrence of dry deposition, there was a slightly positive correlation with coniferous tree species, indicating the occurrence of a filtering effect, which is known to be higher for coniferous trees.

A separate multiple regression analyses including the stand characteristics as well (35 sites) did not indicate a very clear impact of those characteristics on the deposition of S and N compounds. With respect to the deposition of base cations, there appeared to be a significant positive correlation with the stand density index, indicating that more densely populated forest do filter more deposition from the atmosphere. The dataset is, however, too small for a reliable estimate of the various effects. In general, it is well known that e.g. an increased stand height has a positive influence on atmospheric deposition.

## 6.4 Conclusions

Atmospheric deposition data at the Intensive Monitoring plots have been evaluated in view of (i) the relative contribution of wet and dry deposition in the potential acid input, (ii) the relative contribution of N and S compounds and of base cations in the atmospheric input, (iii) the order of magnitude of atmospheric inputs in view of critical loads and (iv) relationships between atmospheric deposition and environmental factors. Major conclusions related to those aspects are given below.

### *The relative contribution of wet and dry deposition in the potential acid input*

The median ratio of the calculated total deposition compared to bulk deposition was approximately 1.6 for both the S and N compounds. On average, the contribution of dry deposition thus appears to be one-third of the total deposition. This is, however, likely to be an underestimate, since bulk deposition partly includes dry deposition (approximately 15-20%, depending on the compound considered). Ninety percent of the ratios ranged between approximately 0.8 and 2.8 for S and between 0.6 and 3.2 for N. Comparatively low ratios were observed in the Nordic countries, where wet deposition appears to be most important, whereas high ratios were found in Western and Central Europe, where dry deposition plays a relative important role.

The ratio of the calculated total deposition of base cations compared to bulk deposition varied in 90% of the plots between approximately 0.46 and 2.76 with a median value of 1.47. This is comparable to the ratio for S and N compounds. However, this result was obtained while excluding a significant number of plots where the ratio of the input of Na by throughfall (measured) and stemflow compared to bulk deposition appeared to be lower than 1.0. Including those plots led to a mean value of approximately 1.33 with 90% of the ranges varying between 0.42 and 2.46. This is clearly lower than for the S and N compounds. This implies that the contribution of base cations, neutralising the acid input is lower in the total deposition than in bulk deposition. As with the S and N compounds, the contribution of wet deposition was dominant in Northern Europe, whereas dry deposition was relatively important in parts of Western and Central Europe.

### ***The relative contribution of N and S compounds and of base cations in the atmospheric input***

On average, the N input in bulk deposition and throughfall equalled the S deposition. The average N to S ratio in the calculated total deposition was, however, nearly 1.5, ranging between approximately 0.5 and 2.7. Even though this result may be influenced by the calculated N uptake rates, this result implies that N is a dominating factor in the acidic input in large parts of Europe. The relative contribution of  $\text{NH}_4$  and  $\text{NO}_3$  in N deposition varied largely over the plots, but in most countries, especially in Northern and Central Europe,  $\text{NH}_4$  seems the dominating N compound at most of the plots. The relationship between Cl corrected base cation deposition and the sum of N and S deposition at the Intensive Monitoring plots is quite low, especially for bulk deposition. A much more significant relationship was observed between the deposition of Ca and  $\text{SO}_4$  both in bulk and total deposition. The correlation may partly be due to associated emissions of  $\text{SO}_2$  and Ca from smelters and refineries (coal burning) and co-deposition of Ca and  $\text{SO}_4$  ( $\text{CaSO}_4$  crystals on stomata).

### ***Ranges and geographic variation of atmospheric inputs in view of critical loads***

Total deposition of S and N compounds ranged between 100-3000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at approximate 90% of the plots, but values up to 4000 and 8000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  were also observed for N and S, respectively. Both bulk and total deposition of N generally appeared to be higher than S deposition at plots in Western Europe (UK, Belgium, Netherlands, Luxembourg, France), whereas the reverse was generally observed at plots in Central Europe (Poland, Czech Republic, Austria, Hungary). For the base cations, the range in total deposition values were more comparable to bulk deposition than to throughfall (mostly between 100 and 2000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  for the sum of Ca, Mg and K) indicating the less significant contribution of dry deposition compared to S and N compounds. The geographic pattern of bulk deposition of Ca, being the most important base cation neutralising the acid input from the atmosphere is quite comparable to S and N, being low in Northern Europe, high in Central and Southern Europe and Intermediate in Western Europe. In Central and Southern Europe the acidic input is thus largely set off by the input of base cations.

Approximately 45% of the considered plots received a N input above 1000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ , being a deposition level at which the species diversity of the ground vegetation may be at risk. Below this deposition level, tree growth may, however, be inhibited. Critical loads related tree health are higher and deposition levels vary from approximately 1000-3500  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ . A comparison with present loads shows that those impacts most likely occur at several plots. The total input of acidity, being the input of S and N compounds minus the deposition of accompanying base cations corrected for Cl, ranged mostly between 200-4000  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ . Considering a variation of critical acid loads related to elevated  $\text{Al}/(\text{Ca}+\text{Mg}+\text{K})$  ratios of approximately 1500-3500  $\text{mol}_c\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ , impacts are likely at part of the plots, but a specific comparison of present and critical acid loads is needed to assess the risk of the acid atmospheric input.

### ***Relationships between atmospheric deposition and environmental factors.***

Geographic region appears to have a dominant influence on the limited data set of deposition data in combination with the various environmental factors. This follows from the results of both a principal component analyses and a multiple regression analyses. Atmospheric deposition of all ions is significantly lower in the Boreal regions compared to Western Europe, whereas  $\text{SO}_4$ ,  $\text{NO}_3$  and Ca deposition is significantly higher in the Central/Eastern part of Europe. There is furthermore a highly significant positive correlation of atmospheric deposition and rainfall, except

for  $\text{NH}_4$  and K. The deposition of S and N compounds appears to decrease significantly with an increase in altitude. For base cation deposition, the impact of altitude is, however, insignificant. A specific point of attention is the calculation of total deposition using a canopy budget model. This requires reliable bulk deposition and throughfall data for all major ions in solution including Na. More emphasis on this topic is relevant for the coming years.



## 7 Meteorological parameters

### 7.1 Introduction

The meteorological data set serves many purposes in the Intensive Monitoring Programme; meteorological data are needed to e.g. compute leaching of substances from soils or to construct input-output budgets for the Intensive Monitoring plots. Eventually, meteorological stress parameters will be included in the analysis of the relation between crown condition and stress factors. This analysis should reveal whether the effects of meteorological extremes on crown condition (additional to effects of e.g. air pollution and unfavourable stand conditions) play an (important) role at the Intensive Monitoring plots.

Unfavourable meteorological conditions can be a serious cause of stress for forest trees, and affect forest condition. Besides negative aspects, meteorological condition can also cause positive effects. Meteorological stress factors include drought stress, temperature stress (cold, frost, heat), radiation stress (lower level of global radiation than the potential level) and mechanic stress (storms, snow, glazed frost). The available data for the Intensive Monitoring plots for 1996 only allow us to focus on temperature stress and drought stress.

Both low and high temperatures can cause stress. High temperatures mainly affect transpiration rates and activity of enzymes. Low temperatures can cause damage in cases of severe or incessant winter frost through freezing or dehydration of needles and buds by which they can be damaged or die off. Late night-frosts in spring can cause severe damage or die off of just flushing buds (Hellings, 1983). Water stress is considered to be very important with respect to forest condition. Innes (1993) mentioned that the most alarming and frequent observations of a decrease in forest condition in Central Europe coincided with the dry years 1982 and 1983. Landmann (1995) mentioned that defoliation appears to be highest in soils poorly supplied with water and/or in stands in which trees, at some stage of development, have suffered from competition for water. The effects of water stress may diverge from yellowing of the foliage, foliage necrosis, to complete defoliation following extreme drought events (Innes, 1993; Landmann, 1995).

Klap et al. (1997) defined various key parameters related to low temperatures (winter index, late frost), high temperatures (heat index) and drought (relative transpiration) in view of their possible impact on the crown condition of pine, spruce, oak and beech forests in Europe. They carried out a statistical analysis that linked defoliation data at the plots of the systematic grid (the so-called level I programme) to stand and site characteristics, air pollution and these meteorological parameters. Results only showed a significant impact of drought stress, in terms of relative transpiration, on the defoliation of oak stands. Similarly, Callaert et al. (1997) give a number of key parameters for low temperatures, drought and heat and excessive wetness that could influence the vitality of pine and beech forest. They carried out a statistical analysis in which they linked defoliation data from pine and beech forests in Belgium to these key factors. They concluded that periods with drought have a negative effect on the vitality of pine forests in Belgium, whereas sufficient precipitation in spring has a positive effect. High temperatures during the vegetation period negatively influenced vitality of both beech and pine. Low winter temperatures positively influenced crown condition of pine, but the reason for this effect is not clear.

The aim of this chapter is to present information on the overall range and variability of calculated natural (meteorological) stress factors. The data assessment methods and the data evaluation methods (the models and their input data) that were used to calculate site-specific meteorological stress factors are described in Section 7.2. A distinction was made in factors indicating temperature stress (winter index, late frost, heat index and summer index) and water stress (relative transpiration). In Section 7.3, the results in terms of the overall range and extremes are presented.

## **7.2 Methodological aspects**

### **7.2.1 Locations**

Figure 7.1 shows the locations of the plots for which countries have indicated that meteorological measurements are (or will be) carried out. The number of plots with stored meteorological data for the year 1996 equals 51. Those plots were located in Finland, Denmark, UK, Ireland, Belgium, Luxembourg, France and Greece. Because most countries started the meteorological measurements only recently, the number of plots for which no data have been stored yet is relatively high. A few countries submitted data that have not been stored yet because of problems with data consistency and/or -quality. These problems will probably be solved this year. For each plot with stored data, the map shows how many parameters have been submitted. The maps shows that for some plots only one or two parameters were submitted (mainly precipitation data) whereas for other plots all mandatory parameters were submitted.

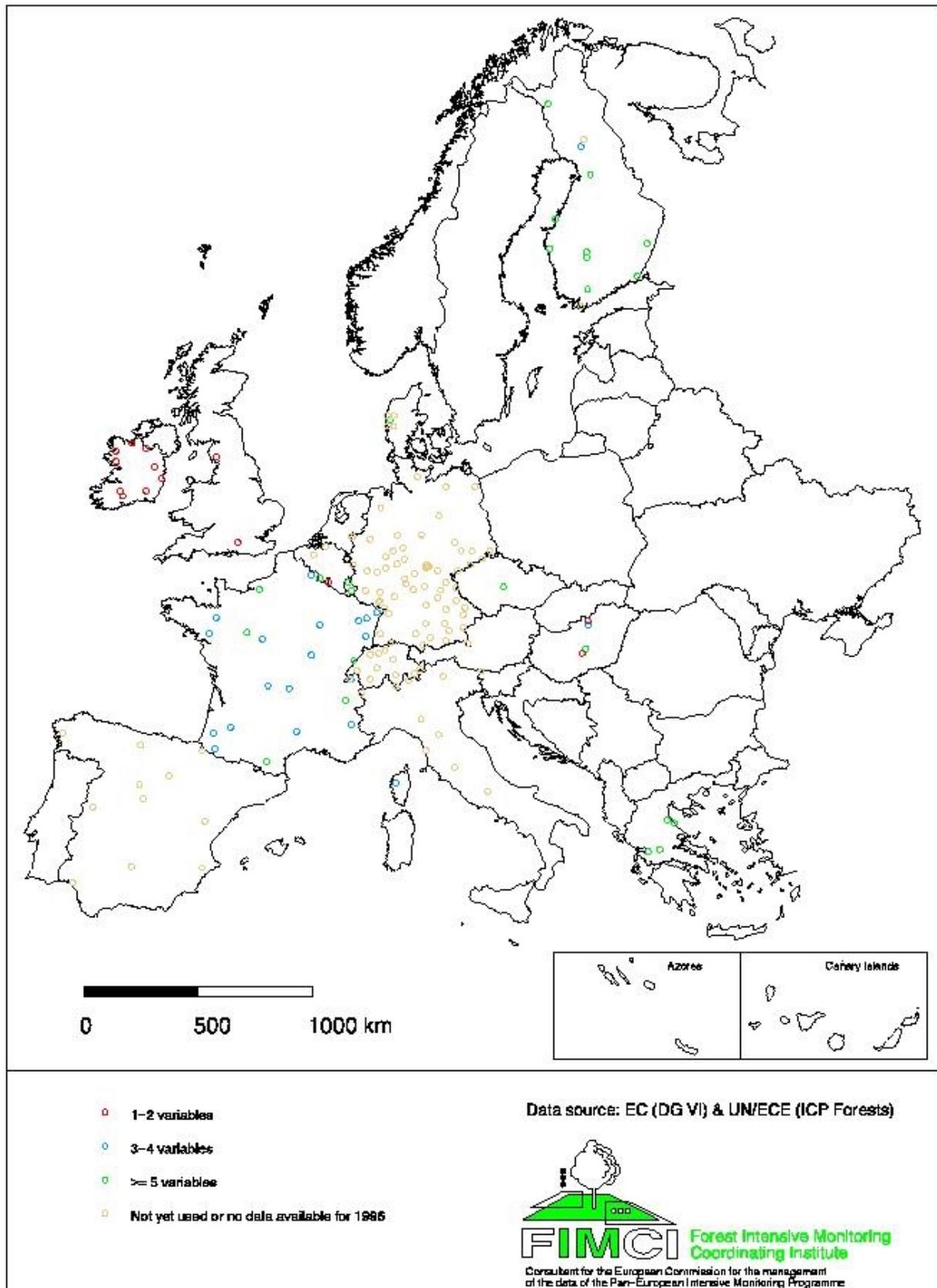
### **7.2.2 Data assessment methods**

Data Accompanying Information on meteorological measurements have been reported for 132 plots, spread over 10 countries. The most relevant information on data assessment methods used for the mandatory data based on a first review of the DAR-Q's is given below. Mandatory data are:

- Precipitation (sum)
- Air temperature (mean, min, max)
- Relative humidity (mean, min, max)
- Wind speed (mean, min, max)
- Wind direction (mean)
- Solar radiation (sum)

#### ***Measuring devices***

Different kinds of equipment are used on the intensive monitoring plots. The majority of countries submitted extensive information on the measuring devices used. Mostly equipment, sensors and their placement has been in accordance to the World Meteorological Organization Standard. For the evaluations carried out in this report, it is assumed that no significant deviations in the measurements have occurred due to measuring errors in the equipment itself.



**Figure 7.1** Geographical distribution of the plots of the Pan-European Intensive Monitoring programme and the number of measured parameters in the meteorological survey in 1996

### **Data collection**

Data collection and storage of the meteorological data was nearly always carried out in a digital way. Only for 4 plots it was reported that measurements were collected and stored in the field solely on paper. Mostly memory cards are used to store the data (63-66% of the plots, depending on the measurement). Data from the memory cards are either transmitted by telephone lines or radio connections, copied in the field or collected by exchanging memory cards.

### **Location of measurements**

In order to obtain data that represent the specific climatological conditions in the forests, various locations of the measurement devices can be used. Measurements for almost all mandatory meteorological parameters were mostly carried out on open field stations (Table 7.1), either within the forest area, in close proximity (in general not more than 2 km distance) of the monitoring plot (79-90% of the plots) or above the forest stand canopy (10-18% of the plots). Most of the open field stations are located within a forest area. Measurements in or under the canopy have been reported only occasionally for air temperature and relative humidity measurements. Data comparability of these plots needs to be considered carefully.

Table 7.1 Location of meteorological measurements. (Numbers between brackets refer to measurements that are carried out in addition to measurements at other locations).

Location	Number of plots				
	PR <sup>1)</sup>	AT <sup>1)</sup>	RH <sup>1)</sup>	WS <sup>1)</sup>	SR <sup>1)</sup>
Above canopy	12	20	13	17	14
In canopy	0	1 (+5)	7	0	0
Under canopy	0 (+3)	0 (+19)	1 (+19)	0 (+6)	0 (+6)
Open field in forest area	73 (+3)	64	62	60	59
Open field outside forest area	38	33	33	14	14
No information	0	5	3	3	3
Total	123	123	119	94	90

<sup>1)</sup> PR = precipitation, AT = air temperature, RH = relative humidity, WS = windspeed and SR = solar radiation.

For a limited number of plots measurements are carried out on multiple locations. These measurements are indicated in Table 7.1 between brackets. These additional measurements are mostly carried out under the canopy and offer interesting evaluation opportunities.

### **Precipitation**

Information on precipitation measurements was received for 123 plots. Most precipitation measurements are carried out within 3 meters from the floor level (103 plots), this mostly concerns the open field stations. On 12 plots measurements were carried out above the canopy on heights ranging between 7 and 40 meter. No significant differences due to the height of measurement are to be expected here.

### **Air temperature**

Methodological information on air temperature measurements has been received for 123 plots. Measurements were mostly carried out on open field stations but also measurements above the canopy were reported (20 plots). Heights of measurements for open field plots range from 1 to 3 meters, whereas the height of the measurements above the canopy vary between 11 to 36 meter above ground level. During evaluations it should be taken into account that air temperature (extremes) may vary with height.

On 24 plots air temperature is measured on two locations per plot. Additional measurements are carried out in or under the canopy. Data measured at these locations are not comparable to open field data or above-canopy data, however they may offer extra meteorological information that can be used for additional evaluations.

#### *Relative humidity*

Methodological information was submitted for 119 plots. Mostly measurements are carried out on open field stations (97 plots). For 13 plots relative humidity was measured above the canopy. Heights of measurements for open field plots vary between 1.5 and 3 meters, whereas the measurements above the canopy range approximately from 10 to 35 meters. For 8 plots measurements were done at locations that differ from the prescriptions in the Manual or EU Regulations. On these plots measurements were carried out in or under the canopy. These data will not be comparable to measurements above the canopy or on open field stations.

#### *Wind speed, Wind direction*

Methodological information for wind speed has been submitted for 94 plots. Wind speed is measured either on open field stations (74 plots) or above the canopy (17 plots). Heights of measurements vary between 1.6 and 37 meter. Wind speed depends on factors as height, roughness of the canopy etc. For evaluations, the local situation needs to be taken into consideration when comparing or using these wind speed data.

#### *Solar radiation*

For 90 plots information on solar radiation was reported. The locations of the solar radiation measuring devices are similar to those of wind speed measurements. Obviously, the heights of measurement do differ. Open field measurements are carried out at heights of 1-3 meters, whereas the heights in measuring towers range from 10 to 40 meters. For 6 plots additional measurements were carried out under the canopy.

#### ***Distance of the meteorological stations to the intensive monitoring plots***

All meteorological stations for which DAR-Q Information has been received are located at or in the vicinity of the intensive monitoring plot. The distance between the meteorological plot and the intensive monitoring plot, for meteo stations that are not on the intensive monitoring plot itself, mostly lies within 100 meters with a maximum of 500 meter (Fig. 7.2 ).

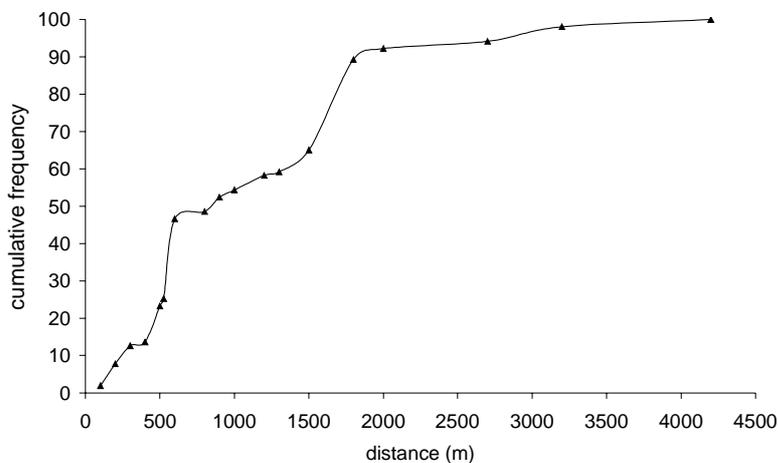


Figure 7.2 Distance of meteorological stations to the intensive monitoring plots.

### 7.2.3 Data quality assurance

#### *Comparison of independent data sets*

Each of the data sets was checked for obvious errors by scanning for values out of plausible ranges. However, because climatic conditions vary strongly over Europe and because plausible ranges must be valid for all countries, plausible ranges are relatively broad and might thus still allow erroneous values in the data sets.

To obtain an indication of the consistency of the data sets, one can compare the sum of daily precipitation from the meteorological data set with the total quantity of bulk deposition from the deposition inventory. These two parameters should approximately have the same value. For about 20 plots, both sufficient data for bulk deposition quantity and precipitation were available to make such a comparison. Results (Figure 7.3) show that the parameters have a high comparability. For 70% of the plots the deviation is less than 5%; only occasionally 10% deviation is found. This means that for plots where no meteorological measurements are carried out, yearly precipitation sums can probably be derived from the deposition data sets without introducing large errors. It must be realised, however, that the frequency of deposition measurements is much lower (generally once every two to four weeks) than the frequency of the meteorological measurements (each day). This means that for computations based on daily values, the deposition data sets cannot be used.

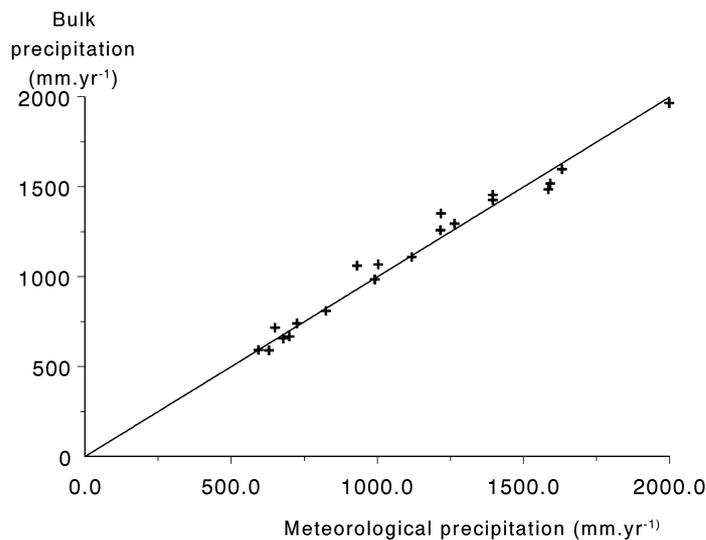


Figure 7.3 Relationship between precipitation based on bulk deposition data and meteorological data.

#### *Use of external data bases*

Although not part of the quality assurance of the monitoring data, external meteorological databases that may be used in several evaluations, should also be reviewed in terms of consistency and reliability. To derive, for example, relationships between deposition or soil solution and environmental characteristics, meteorological parameters (specifically precipitation data) are often relevant (see chapters 6 and 8). For plots where neither meteorological measurements nor deposition measurements are made, precipitation may be derived from external data sources. Such a data source is e.g. given by Leemans and Cramer (1991) who interpolated

long term average monthly meteorological data from about 2000 meteorological stations in Europe to a 0.5 x 0.5 ° grid system.

A comparison between measured 1996 precipitation data of 23 intensive monitoring plots and the long term precipitation from the associated grid cell showed, in general, a reasonable accordance (Figure 7.4). For 80% of the plots the difference was less than 25%. Only for a few plots strong deviations occur (up to a factor 2.5). Differences can occur because data from one specific year (1996) are compared to long term average data which can lead to strong deviations. Furthermore, an Intensive Monitoring plot can be located at a location with circumstances (such as altitude or exposure) that strongly deviate from the ‘average’ circumstances in the associated grid cell. As a first indication of the ‘average’ value, grid data for precipitation may be used, but one should be very careful in using these data when values from a specific year are needed.

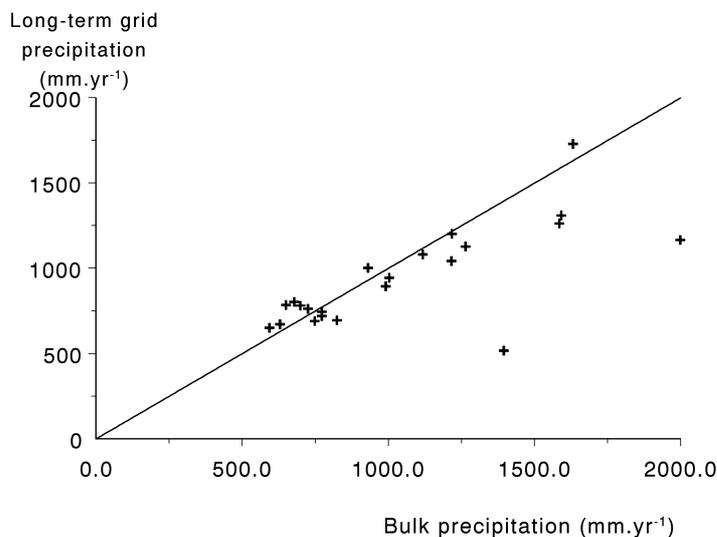


Figure 7.4 Comparison of long-term average precipitation data derived from external databases and precipitation data for the year 1996 based on bulk deposition measurements.

From section 7.2.2 it becomes clear that measurements of meteorological parameters are made at various locations (open field, in the canopy, above the canopy). For at least part of the parameters, there are possibilities to make the values comparable, using equations that describe the parameter value as a function of e.g. height. This will be elaborated further in co-operation with the chairmen of the expert panel on meteorology.

## 7.2.4 Data evaluation methods

### 7.2.4.1 Assessment of temperature stress indices

#### *General approach*

As indicated in section 7.1, meteorological conditions during the growing season can have important effects on the condition and damage of the forest ecosystem. Several authors have given key-parameters that relate to meteorological stress (e.g. Klap et al., 1997, Callaert et al., 1997).

With respect to the temperature stress the following key parameters were given by Klap et al (1997):

- Winter index: indication of severeness of the winter. This index equals the sum of daily mean temperatures below 0 °C in the period from 1 October to 1 April (degree-days below 0 °C).
- Late frost: an indications of the severeness of late night-frost in spring that can cause serious damage to trees when growth has just started and the buds and young shoots are very sensitive to frost. This index is defined as the lowest minimum temperature (below 0 °C) in a period starting 15 days before the beginning of the growing season and ending at June 30.
- Heat index: an indication of the possible occurrence of damage by high temperatures, computed as the sum of differences between daily maximum temperatures in the growing season and a (preliminary) threshold value of 35 °C (degree days above 35 °C).
- Summer index: an indication of the quality of the growing season, as it affects the possibilities of photosynthetic activity and the possibility of the tree to produce reserve assimilates for purposes of defence and growth in the beginning of the next season. This index can be calculated as an effective temperature sum, which equals the sum of differences between daily mean temperatures during the growing season and a (preliminary) threshold of 5 °C (degree-days above 5 °C).

As these parameters are based on (daily) temperature data alone, they can be computed for sites where mandatory data have been collected, especially because temperature is a parameter that is only very occasionally lacking in the mandatory data sets. For the evaluation of this year, results are presented for late frost and summer index only. When in later years meteorological stress is related to other forest ecosystem characteristics, also the other parameters (winter index and heat index) will be taken into account.

On request of (amongst others) the European Commission, Callaert et al. (1997) carried out a study in which they present an literature overview of damage to *Pinus sylvestris* and *Fagus sylvatica* by meteorological extremes. They give a number of additional key parameters for low temperatures (e.g. number of days from October to May with temperatures below -4 °C), drought and heat (e.g. total number of days in the vegetation period (with a minimum of 5 days) without rainfall), and excessive wetness (e.g. total number of days in the vegetation period (with a minimum of 5 days) with a rainfall of more than 3 mm). Some of these key parameters are probably also useful for studies on a European scale, and will be included in future evaluations that include both crown condition and meteorological data.

### ***Assessment of the growing season***

Combination and aggregation of meteorological data over periods where the ecosystem is vulnerable for damage can give information on a-biotic damage by unfavourable meteorological conditions. For such assessments, the start and end date of this vulnerable period (that depend on climate and species) must be known. At the moment the ad-hoc working group on phenology is elaborating a proposal to determine the start and end dates of the vulnerable periods on plot level. Until the results of this study become available, the start and end of the growing season were set to fixed dates, depending on bio-geographic region and tree species. Table 7.2 shows the dates for the beginning of the growing season, based on literature data (Klap et al., 1997b).

Table 7.2 Start of the growing and dormant season per tree species for climatic regions

Climatic region <sup>1)</sup>	Starting date growing season			Starting date dormant season (all species)
	<i>Q. petraea</i>	<i>P. sylvestris</i> <i>Q. robur</i> <i>Q. ilex</i>	<i>P. abies</i> <i>F. sylvatica</i>	
Boreal (Lat. > 65° N)	May 1	May 16	June 1	September 16
Boreal (Lat. < 65° N)	April 16	May 1	May 16	September 16
Boreal Temperate	April 1	April 16	May 1	September 16
Mountainous N. (Alt. > 500m or Lat. > 65° N)	April 16	May 1	May 16	September 16
Mountainous N. (Alt. < 500m and Lat. < 65° N)	April 1	April 16	May 1	September 16
Sub-Atlantic	March 16	April 1	April 16	October 1
Continental	March 16	April 1	April 16	October 1
Atlantic North	March 16	April 1	April 16	October 1
Atlantic South	March 1	March 16	April 1	October 16
Mountainous South (Alt. < 1500 m)	March 16	April 1	April 16	October 1
Mountainous South (Alt. > 1500 m)	April 1	April 16	May 1	October 16
Mediterranean Lower	Feb. 15	March 1	March 16	October 16
Mediterranean Higher	March 1	March 16	April 1	October 16

<sup>1)</sup> Based on the regions used in the annual Forest Condition Reports (e.g. UN-ECE, EC, 1996).

The beginning of the growing season first starts in the southern regions and then moves northwards, whereas the end of the growing season has the opposite trend. The beginning of the growing season was considered to be species-dependent. *Quercus petraea* was considered to be an 'early' species, whereas *Fagus sylvatica* and *Picea abies* were considered to be 'late' species.

#### 7.2.4.2 Assessment of drought stress indices

##### General approach

With respect to drought stress, relative transpiration may be a suitable stress factor. It can be calculated as the ratio between actual transpiration and potential transpiration ( $E_{t_{act}}/E_{t_{pot}}$ ) (Klap et al., 1997). One of the best known examples of a deterministic formula to estimate potential evapotranspiration is the formula of Penman/Monthieith (Monthieith, 1965). It has been successfully applied in many studies. The drawback is its complexity: it needs many input parameters for computation of the potential evapotranspiration.

The equation is given as:

$$\lambda E_t = \frac{\delta R_n + \rho * C_p * (e_s - e)/r_a}{\delta + \gamma * (1 + r_s/r_a)} \quad (7.1)$$

With:

- $\lambda E_t$  = Latent heat loss in evapotranspiration ( $J.m^{-2}.s^{-1}$ )
- $\delta$  = Slope of the saturation vapour pressure-temperature curve ( $mbar.^{\circ}C^{-1}$ )
- $\gamma$  = Psychrometer coefficient ( $mbar.^{\circ}C^{-1}$ )
- $R_n$  = Net radiation ( $W.m^{-2}$ )
- $\rho$  = Density of dry air ( $kg.m^{-3}$ )
- $C_p$  = Heat capacity ( $J.kg^{-1}.^{\circ}C^{-1}$ )
- $e_s$  = Saturated vapour pressure (mbar)
- $e$  = Vapour pressure at temperature  $T_a$  (mbar)
- $r_a$  = Aerodynamic resistance ( $s.m^{-1}$ )
- $r_s$  = Stomatal resistance ( $s.m^{-1}$ )

Where  $\gamma$  equals  $0.67 \text{ mbar} \cdot \text{°C}^{-1}$ ,  $\rho$  is  $1.2047 \text{ kg} \cdot \text{m}^{-3}$  and  $C_p$  equals  $1004 \text{ J} \cdot \text{kg}^{-1} \cdot \text{°C}^{-1}$ . Net radiation can be computed from measured global radiation, saturated vapour pressure and the slope of the saturation vapour pressure-temperature curve from temperature, and actual vapour pressure from measured relative humidity and saturated vapour pressure. The aerodynamic resistance can be computed with various equations ranging from simple to complex, but the wind speed is normally part of these equations. This means that for the use of the Penman/Monteith equation, all mandatory parameters (except for precipitation) are needed. It can thus only be applied to those sites where these parameters are available throughout the year.

To compute the ratio between actual and potential evapotranspiration, the actual evapotranspiration must also be computed. Then, information is needed on transpiration and evaporation reduction as caused by inadequate water supply. Often, transpiration reduction is related to soil moisture depletion (see e.g. Doorenbos and Kassam, 1979) or soil matric head. This means that a soil water balance needs to be computed, which, in turn, requires information soil physical characteristics. Many models exist that simulate the soil water balance, ranging from simple bucket models (see e.g. Kalma et al., 1995) to complex non-stationary deterministic models (see e.g. Kabat et al., 1992). As the availability of soil physical data in the Pan-European Monitoring Programme is very limited (only voluntary soil texture data are available) and the number of computations quite numerous, the data demand and complexity of the used model should not be too high.

Because of its expected high accuracy, the Penman/Monteith equation for potential evapotranspiration was used for the assessment of drought stress. It was combined with a simple model for the simulation of soil water storage and depletion (Klap et al., 1997). In the model described by these authors, interception is computed according to a procedure described by Gash et al. (1995). Transpiration reduction caused by insufficient water supply is computed according to Doorenbos and Kassam (1979, see above). The data demands of this model limited the number of sites to which it could be applied (for 1996) to 12. Apart from the meteorological data, data on soil and stand characteristics were needed. Soil characteristics were derived from soil type (available water capacity) and the soil survey (C and N contents of the topsoil). The stand characteristics, being the number of trees per hectare and tree species, were derived from general plot data. Tree height was estimated using an improved version of the procedure described in Klap et al. (1997), that relates tree height to tree species, stand age and site quality. In principle tree height can be derived from the increment survey, but for most of the 12 plots these data were not yet available. The estimate of the available water capacity which is part of the site quality can also be improved in the future by relating it not only to soil type but also to the voluntary data on soil texture.

### ***Possible alternatives***

The data need of the applied model is fairly high and, as a consequence, its applicability is limited. As an alternative one could use an empirical relationship to derived potential evapotranspiration that needs less input data and can thus be applied to more plots. An example of such an empirical formula is the well-known Thornwaite equation (Thornwaite, 1954):

$$Et = 1.6b(10t/L)^a \quad (7.2)$$

With

$E_t$  = monthly potential evapotranspiration (cm)

$t$  = mean monthly temperature ( $^{\circ}\text{C}$ )

$a, b, L$  = empirical parameters

where  $L$  can be calculated from the mean monthly temperatures  $t$ :

$$L = \frac{12}{1} (t/5)^{1.514} \quad (7.3)$$

Potential transpiration is thus linked to temperature alone, whereas in reality it is dependent on more parameters such as humidity, wind speed and radiation. These factors are implicitly incorporated in Thornwaite's method by assuming relationships between e.g. temperature and relative humidity and between temperature and radiation. This however, implies that this formula does not give reliable results in conditions with rapid changes in temperature as compared to e.g. relative humidity and radiation. Despite its simplicity it has proven to give reasonable results in almost every climate area in the world (see Ward, 1974).

In case no data on soil characteristics are available, one could also consider using the difference between precipitation and potential evapotranspiration as the simplest indicator of drought stress. If the precipitation (on a weekly or monthly basis) is lower than the potential evapotranspiration, one can assume that transpiration will be reduced in that period. The advantage of this indicator is that it can be computed without any modelling of soil water balances; this disadvantage is of course that it does not take into account water storage in soils or capillary rise. It can therefore only be used as a rough indicator of drought stress and not as a measure of the water balance of the ecosystem. Some further research is needed in the future to see which of the methods is most appropriate to estimate drought stress, from the point of both accuracy and applicability.

## 7.3 Results and discussion

### 7.3.1 Temperature stress indices

The key parameters late frost and summer index were calculated for 35 plots, where sufficient data were available for 1996. These plots are located in Finland, Denmark, Belgium, UK, France and Greece. Since the number of plots was so limited, they do not give an overall European picture.

Late frost (Figure 7.5A), computed as the lowest minimum temperature (below  $0^{\circ}\text{C}$ ) in a period starting 15 days before the beginning of the growing season and ending at June 30, is strongly dependent on location. Lowest temperatures are of course found in Northern Europe but because the growing season starts late in these regions, late frost might not occur. Late frost in 1996 occurs on 25 of the 32 plots in 1996, the minimum temperature is generally between  $0$  and  $-5^{\circ}$ . The most severe late frost was recorded for a high altitude plot (900-950m) in central France ( $-8.2^{\circ}\text{C}$ ). For the plots in Greece, 3 plots in France and 1 plot in Finland, no late frost occurred.

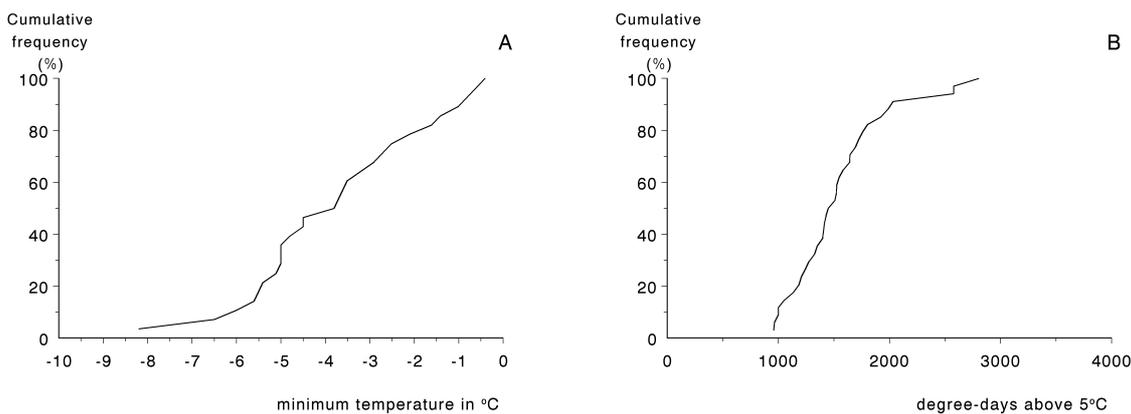


Figure 7.5 Cumulative frequency distributions of late frost (A) and summer index (B) at 32 Intensive Monitoring plots.

The summer index (Figure 7.5b) was computed as an effective temperature sum, which equals the sum of differences between daily mean temperatures during the growing season and a (preliminary) threshold of 5 °C (degree-days above 5 °C). This factor is thus also affected by the estimated growing season: it will be lowest in northern Europe (low temperatures and a short growing season) and highest in the Mediterranean area (high temperatures and a longer growing season). Results show that the summer index varied between less than 1000 degree-days above 5 °C in Northern Europe (Finland, Denmark and the northern part of the UK) and high altitude plots in France, to about 2800 degree-days above 5 °C in Greece. The summer index has no direct link to forest vitality, but is an indication of the quality of the growing season. As such it probably has an effect on the forest growth at the various plots. This should be investigated when in the future forest growth data are linked to meteorological data.

### 7.3.2 Drought stress indices

Because the model that computes the drought stress needs all mandatory meteorological data for a whole year, the model could only be applied to 12 intensive monitoring plots, located in France, Luxembourg and Greece. For the other plots, required meteorological parameters were missing, measurements were available for only part of the year or meteorological parameters were measured only in or above the canopy. Although measurements above the canopy are in accordance with the Intensive Monitoring manual, these plots were presently not considered because of possible incomparability of data. More information on comparability and re-scaling of parameters measured in or above the canopy is needed before these data can be used to compute drought stress (cf section 7.2.2).

Because some of the input parameters were estimated using transfer functions, the results from the calculations have a considerable uncertainty. The only validation that could be carried out at this point was a comparison of computed throughfall (rainfall minus interception evaporation) and measured throughfall (sum of throughfall quantities from the deposition inventory). This comparison gives a first indication of how well the interception module in the model simulated the actual throughfall, and could be made for 8 of the 12 plots where actual throughfall data for the whole year were available from the deposition survey. Results of this comparison show that the actual and simulated throughfall compare well; for 6 of the 8 plots the difference was less than 15%, for the other 2 plots the deviation is between 20 and 30%. The difference in computed

interception and measured interception (rainfall minus throughfall) was relatively larger (Table 7.3).

*Table 7.3 Comparison of measured and calculated throughfall and interception evaporation at eight Intensive Monitoring plots.*

Country	Plot no.	Throughfall (mm/yr)		Interception evaporation (mm/yr)	
		Measured	Calculated	Measured	Calculated
France	37	718	626	212	304
	75	497	478	125	144
	86	388	493	261	156
	93	1175	1112	341	404
	94	1164	1039	263	388
	96	1112	1139	409	382
Greece	2	1106	1155	220	171
	4	1038	1337	722	423

Computed potential transpiration varied between 440 and 710 mm, whereas actual transpiration varied between 240 and 440 mm for 1996. Computed transpiration reductions (reduction of potential evapotranspiration) varied between 34% in Luxembourg to about 60% for one of the Greek plots. Computed precipitation surpluses varied between 150 and 1000 mm. The information on the precipitation surplus, being the drainage from the rootzone to groundwater, is crucial to calculate the element output to groundwater. Precipitation surplus is not a useful key parameter for drought stress. For example, the highest precipitation surpluses were found for some plots in Greece and France, that also have high transpiration reductions. On these plots, most of the high amount of annual precipitation falls in the wintertime. In the summer, rainfall on these plots is low so substantial transpiration reduction can still occur.

The computations performed with the 1996 meteorological data have a tentative character. The available data set was very limited and the simulation of drought stress could only partly be validated. In the coming years the data set will expand and drought stress simulations may be improved by more accurate parameters estimates, both for input parameters and for model parameters. Wherever possible, the simulation results should be validated. Furthermore, simpler methods for computations of drought stress (described in section 7.2) should be evaluated that are less data demanding and can therefore be applied to more of the Intensive Monitoring plots. Finally, it is necessary to investigate the possibilities to re-scale meteorological measurements made at various heights to standard height.

## 7.4 Conclusions

The available meteorological data did allow the calculation of key parameters, such as to low temperatures (winter index, late frost), high temperatures (heat index) and drought (relative transpiration) in view of their possible impact on crown condition and forest growth. Late frost, computed as the lowest minimum temperature (below 0 °C) in a period starting 15 days before the beginning of the growing season and ending at June 30, occurred on 25 of the 35 plots where sufficient data were available for 1996. These plots were located in Finland, Denmark, Belgium, UK, France and Greece. Lowest temperatures were of course found in Northern Europe but because the growing season starts late in these regions, late frost might not occur. The summer index, computed as the sum of differences between daily mean temperatures during the growing season and a threshold of 5 °C (degree-days above 5 °C) varied between less than 1000 degree-

days above 5 °C in Northern Europe (Finland, Denmark, the northern part of the UK and high altitude plots in France, where are temperatures low and the growing season is short), to about 2800 degree-days above 5 °C in Greece (high temperatures and a longer growing season).

Because the model that computes the drought stress needs all mandatory meteorological data for a whole year, the model could only be applied to 12 intensive monitoring plots, located in France, Luxembourg and Greece. Computed potential transpiration varied between 440 and 710 mm, whereas actual transpiration varied between 240 and 440 mm for 1996. Computed transpiration reductions (reduction of potential evapotranspiration) varied between 34% in Luxembourg to about 60% for one of the Greek plots. The quality of the calculated drought stress indices depends on the quality of the model and the data. Validation was only possible for the interception module of the hydrological model, by comparing computed interception (rainfall minus throughfall) and measured interception at 8 of the 12 plots where all data were available. Results of this comparison showed that the actual and simulated interception compare reasonably well for 6 of the 8 plots (a difference of approximately 5 – 30%). For the other 2 plots the deviation is larger. An indication of the quality of the precipitation data was obtained by evaluating the consistency of the sum of daily precipitation from the meteorological data set with the total quantity of bulk deposition from the deposition inventory at 20 plots, where sufficient data were available for such a comparison. For 70% of the plots the deviation was less than 5%; only occasionally 10% deviation was found. This means that yearly precipitation sums seem quite reliable.

The computations performed with the 1996 meteorological data have a tentative character. The available data set was very limited and the simulation of drought stress could only partly be validated. In the coming years the data set will expand and drought stress simulations may be improved by more accurate parameters estimates, both for input parameters and for model parameters. Wherever possible, the simulation results should be validated. Furthermore, simpler methods for computations of drought stress (described in section 7.2) should be evaluated that are less data demanding and can therefore be applied to more of the Intensive Monitoring plots. Furthermore, it is necessary to investigate the possibilities to re-scale meteorological measurements made at various heights to standard height.

## 8 Soil solution chemistry

### 8.1 Introduction

In general, soil solution chemistry has a fast response to changes in atmospheric deposition. This is specifically true for elements with few interactions with the mineral soil, such as Cl and SO<sub>4</sub>. In most cases, both ions act as a tracer but SO<sub>4</sub> adsorption does occur, especially in the B horizons of podzolic soils. For other elements, such as NO<sub>3</sub> and Al, the chemical soil composition strongly affects the soil solution chemistry. For such elements, the concentration in the soil solution is mainly a result of atmospheric deposition, hydrology and the interaction with the mineral soil.

This chapter focuses on the chemistry of major ions in soil solution impacted by N and S deposition, either directly (SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>) or indirectly through soil buffering reactions (H, Al, Ca, Mg, K). Apart from the presentation of the range in ion concentrations at various soil depths by means of cumulative frequency distributions, results are also evaluated in view of available critical levels in the literature. This refers to:

- NO<sub>3</sub> in view of N saturation and groundwater pollution
- Al in view of acidification and groundwater pollution
- Ratios of NH<sub>4</sub> and Al to base cations in view of nutrient imbalances

Specific emphasis is, however, given to relationships between (i) element concentrations in soil solution and soil characteristics (e.g. Al concentration or pH versus base saturation) (ii) concentrations of various elements in soil solution (e.g. Al vs. pH or Al versus SO<sub>4</sub>+NO<sub>3</sub> in acid soils) and (iii) element concentrations in soil solution and atmospheric deposition (e.g. SO<sub>4</sub> concentration versus S deposition and NO<sub>3</sub> concentration versus N deposition). Element concentrations are simple (annual) average concentrations, since water flux calculations can not yet be carried out.

Relationships focus specifically on NO<sub>3</sub> and Al and include those between:

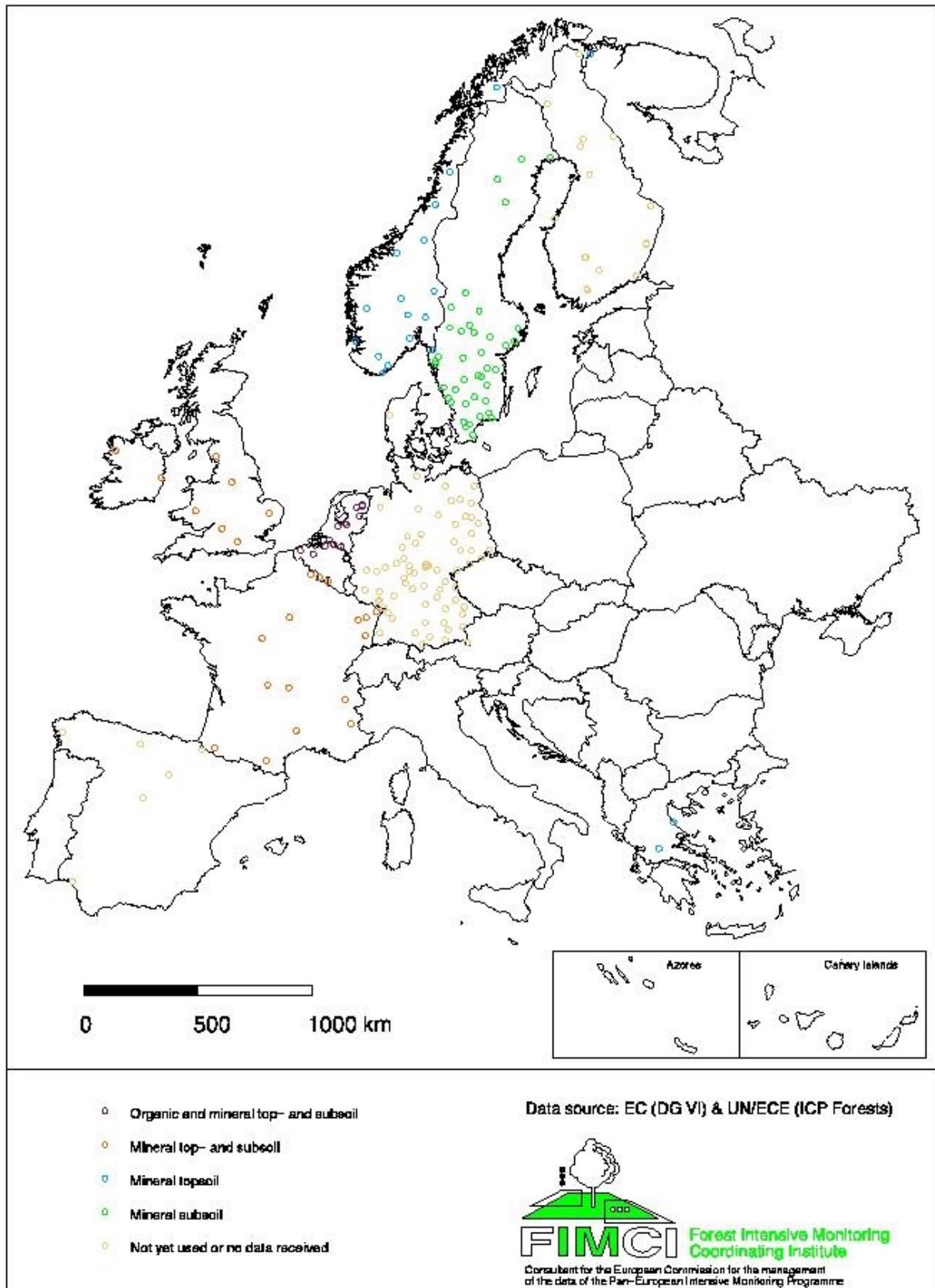
- NO<sub>3</sub> concentration versus N deposition and C/N ratio of the soil (Gundersen et al., 1998)
- Al concentration versus SO<sub>4</sub> and NO<sub>3</sub> concentration (reflecting inputs of S and N compounds) and the base saturation of the soil (De Vries et al., 1995)

### 8.2 Methodological aspects

#### 8.2.1 Locations

Data for the soil solution chemistry in 1996 were stored for a total of 103 plots in eight countries. The total number of countries where soil solution chemistry measurements take place equals 12. Finland and Spain, did, however, not yet submit data, whereas the data submitted by Germany and Denmark did not yet pass the validation process (Fig. 8.1). Sweden only measured the chemical composition in the subsoil (40-80 cm) whereas Norway and Greece concentrated their measurements in the topsoil (0-40 cm).





**Figure 8.1** Geographical distribution of the plots of the Pan-European Intensive Monitoring programme and layers sampled in the soil solution survey of 1996

U.K. and France measured in both topsoil and subsoil, whereas Ireland, the Netherlands and Belgium measured the chemical composition of the soil solution in the organic layer, mineral topsoil and mineral subsoil (Table 8.1). Apart from Greece, all plots are concentrated in Western and Northern Europe (Fig 8.1.)

Table 8.1 Number of plots at which soil solution chemistry was measured in 1996 in the various participating countries<sup>1)</sup>

Country	Number of plots			
	Organic layer	Mineral topsoil <sup>2)</sup>	Mineral subsoil <sup>2)</sup>	All layers
Sweden	-	-	41	41
Norway	-	17	-	17
U.K.	-	6	6	6
Ireland	3	3	3	3
Netherlands	11	11	11	11
Belgium	6	8	7	8
France	-	15	15	15
Greece	-	2	-	2
All	20	62	83	103

<sup>1)</sup> Numbers refer to the data that were stored and used in the data evaluation.

<sup>2)</sup> Mineral topsoil is 0-40 cm and mineral subsoil is 40-80 cm.

## 8.2.2 Data assessment methods

Methodological information has been submitted and stored for 145 monitoring plots. Also in the submission files for soil solution some information on the applied monitoring approach is given. Submission files have been received for 120 plots although data have only been stored for 103 plots following a thorough data validation procedure. As with meteorological data, the information on data assessment methods for the first results of this survey has been derived from the DAR-Q's and thus apply to 145 plots.

### *Sampling devices*

Soil solution can be collected by various methods. Either soil solution collectors (zero tension lysimeters or suction cups) can be placed or soil solution samples can be obtained by taking soil samples and extraction of soil solution using either centrifugation or extraction methods. Depending on the aim and frequency of the monitoring and the soil condition, use can thus be made of (i) tension lysimetry, (ii) zero-tension lysimetry, (iii) centrifugation and (iv) the saturation extract method. The ion concentrations obtained do depend on the measuring devices, since different types of soil water are extracted. In general, concentrations increase going from zero tension lysimeters to suction cups and to centrifugation. Some information on differences is given in Annex 1, based on a comparative study in Finland (Derome and Lindroos, 1997) and the Netherlands (Verhagen and Diederer, 1991). More in-depth quantitative information on the differences, accounting for differences in the suction cup material, requires a complete review of the available literature on soil solution sampling, complete with consultation with experts in the field and a careful examination of the DAR questionnaires. Such a study is foreseen by Finland and Denmark. Hopefully, results can be included in next years report.

The use of (zero)tension lysimeters is the reference method for soil solution collection. On the majority (89%) of plots, these non-destructive methods have been applied (Table 8.2).

*Table 8.2 Soil solution collection methods applied in the Intensive Monitoring Programme*

Collectors	Number of plots
Tension lysimeters / suction cups	93
Zero tension lysimeters	2
Tension meters/ suction cups + zero tension lysimeters	34
Soil sampling and saturation extraction	2
Soil sampling and centrifugation	14
Total	145

The most appropriate use of (zero)tension lysimeters is strongly depending on local circumstances. Therefore various types of lysimeters are applied. Tension cups are in general preferred since they cause little disturbance in the soil and because they allow sampling in dry summer months when there is no vertical movement of water. Tension plates are the most appropriate for sampling of soil solution below the humus layer. Zero-tension lysimeters are most appropriate for sampling soil solution that is transported through macropores, particularly after precipitation events of high intensity. On almost all of the plots where (zero)tension lysimeters are installed, suction cups are used (97%). On 34 plots combinations of types of samplers are reported. In these cases, suction plates or zero tension cups/plates are applied in addition to suction cups (see also Table 8.3).

The materials used for the lysimeters differ between the various plots. Lysimeters used should be made of materials that are considered sufficiently free of contaminants, such that the sample solution is not influenced by the sampler itself. In the submanual for soil sampling (UN-ECE, 1998) a list of materials considered sufficiently free of contaminants is presented. The majority of used lysimeters is made of materials that are considered appropriate (Table 8.3). Only the materials aluoxide and INOX were not mentioned in the above mentioned list and thus require special attention. Note that Table 8.3 only presents information on the number of plots where lysimeters were applied. For the 16 plots in where soil sampling and centrifugation (11 plots in the Netherlands) or saturation extraction (2 plots in Greece) has been applied, no further information on materials used has been stored.

*Table 8.3 type and construction material of lysimeters used. Numbers between brackets are additional measurements.*

Lysimeter	Material	Number of plots
Suction cup	Ceramic	65
	Teflon	36
	Plastic	2
	Aluoxide	22
Suction plate	Aluoxide	3
	Ceramic	(6)
Zero tension cup	Aluoxide	(3)
Zero tension plate	Plastic	1 (+ 4)
	INOX	(6)
Total		126 (+19)

### ***Sampling numbers***

Due to high spatial variation in both soil solution chemistry and percolation water fluxes, caused by e.g. variation in tree cover, ground vegetation and soil properties, a sufficient number of samples is needed to obtain a reliable estimate of the average concentration on a plot scale. This number increases when the spatial variability is larger (Section 3.2).

On average 10.6 lysimeters per plot are used for the soil solution collection. Figure 8.2 gives a cumulative frequency distribution of the number of samplers used per plot. In this figure all samplers per plot are lumped, regardless of the number of layers sampled per plot. Numbers of samplers per plot range from 1 up to 45, with the majority (70%) varying between 10 and 20 samplers per plot. The average number of layers sampled per plot was 2.2, thus leading to an average of 4.8 replicates per layer.

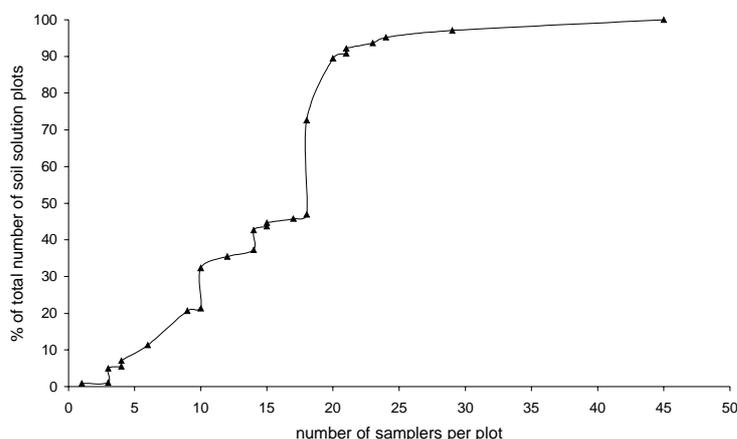


Figure 8.2 Cumulative frequency distribution of the number of samplers (lysimeters and/or zero tension lysimeters) used at the Intensive Monitoring plots

An indication of the adequacy of the number of samplers or samples can be derived from the results of a comparative study of three methods to extract soil solution at two forest stands in the Netherlands at fifteen spots (see also Annex 1). The relative standard deviation of the estimated mean concentration at each depth varied nearly always between 20 and 60% (Table 8.4; see also De Vries and Leeters, 1999).

Table 8.4 The standard deviation relative to the mean concentration of major elements in the soil solution of two Dutch forest stands at four soil depths<sup>1)</sup>.

Depth (cm)	Location	Relative standard deviation (%)									
		H	Al	Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	SO <sub>4</sub>	Cl
10	IJsselstein	52	26	22	40	34	34	62	26	38	32
20	Speuld	44	50	30	40	46	36	68	60	52	32
40	IJsselstein	46	58	42	36	34	20	62	28	32	22
60	Speuld	38	58	80	28	34	34	42	62	36	42

<sup>1)</sup> The number of samples taken at each site equals 15.

The relative standard deviation, which is related to the spatial variability, was relatively small for Na and Cl (mostly between 20 and 35%) and relatively large for Al and NH<sub>4</sub> (mostly between 50 and 60%). Based on the results of this study, the number of sub-samples (in one pooled sample) taken in the soil survey in Intensive Monitoring stands in the Netherlands equals 20. Using this number, the margin of error for most elements is less than 20%. Note that the margin of error, D, is about half the relative standard deviation, S, reported in Table 8.4 (See Eq. 3.2 in Section 3.2 with N = 15 and t<sub>α</sub> = 1.96). This implies that the reliability of the average value generally varies between ± 20%.

Considering the variation in the two Dutch forest plots to be representative, the number of suction cups or zero tension lysimeters generally used seem quite low. Requiring that the number of samples should be such that the plot mean is within ± 20% of the population mean with a

confidence level of 95%, at least 10 samples are needed, assuming a relative standard deviation of 30% (compare Table 8.4 and Table 3.1).

### ***Sampling layout***

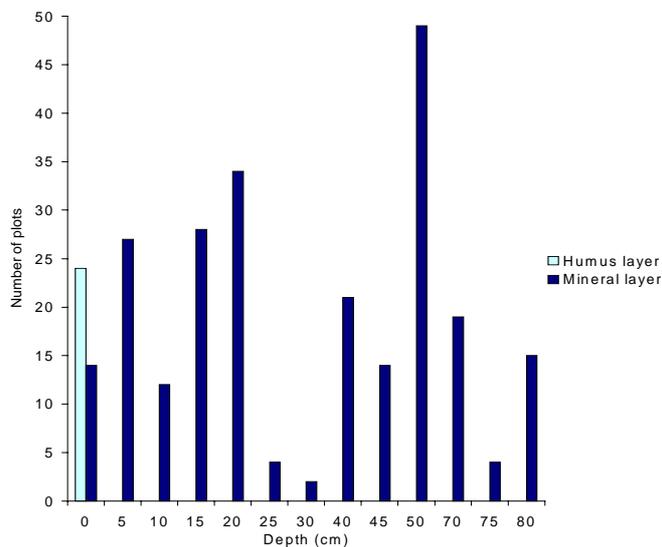
Representativity of the measurements for the plot is furthermore depending on the distribution of the lysimeters over the plot. Samplers are placed either randomly or systematically over the plot. Table 8.5 gives an overview of the various distributions that have been used.

*Table 8.5 Distribution of lysimeters through the plot*

Placement of lysimeters	Number of plots
random	32
random+sampl.grid	1
sampling grid	24
cross-fixed distance	3
line-fixed distance	25
line-random distance	1
other	14
No information	45
Total	145

### ***Sampling depths***

In general there is quite a strong gradient with depth for most of the elements considered. Therefore it is relevant to have information on the various sampling depths. The depths of the sampled layers are presented in Figure 8.3. Mostly soil solution collection has taken place in mineral layers. Measurements in the humus layer or organic layers were reported only for a few plots. The average number of layers sampled was 2.2. Mostly countries carry out one measurement in the topsoil (0-40 cm) and one measurement in the sub-soil (40-80 cm).



*Figure 8.3 Overview of soil solution sampling depths.*

### ***Conservation and analysis of the samples***

Good quality of the soil solution data requires a careful treatment of the sample water. Contamination of the samples by (e.g. algae) is prevented by keeping the samples cool and dark

in the field and cleaning or replacing collection bottles periodically. DAR-Q's report these measures for all plots. Also preservatives are used to diminish biological activity in the sample.

All countries conserve their sample water in cool places. Cooling temperatures range from -20 up to 10 degrees Celsius, however mostly a cooling temperature of 4-6 degrees Celsius has been applied.

The submanual on soil solution sampling (UN-ECE, 1998) states that samples should be analysed as soon as possible on the untreated samples for pH and conductivity. Furthermore filtration (0.45 mm membrane filtration) should be applied. Hereafter at least the other mandatory parameters dissolved organic carbon (DOC), K, Ca, Mg, Al total (if pH<5), NO<sub>3</sub>-N and SO<sub>4</sub>-S and optional parameters should be measured. Based on a first inventory of the DAR-Q information no specific problematic pre-treatment and analysis methods were noticed. However the use of ringtests on a European scale like the ones applied for foliage chemistry and deposition data is needed here as well to improve the insight into the data quality of the various analyses. About half of the countries that have submitted DAR-Q information reported to be involved in such ringtests already.

### 8.2.3 Data quality assurance

As with atmospheric deposition, the procedures for quality assurance and quality control (QA/QC) included a check on:

- the balance between cations and anions
- the difference between measured and calculated electric conductivity
- the ratio between ion concentrations

#### *Ionic balance*

The difference between the sum of all major cations and anions in soil solution was calculated comparable to Eq. (6.1) – Eq. (6.3). However, unlike atmospheric deposition, Al was included in the calculation of cations. Furthermore, when DOC measurements were available, an estimate was made of the concentration of organic anions according to (Oliver et al., 1983):

$$\text{RCOO}^- = m \cdot \text{DOC} \cdot \frac{\text{Ka}}{\text{Ka} + [\text{H}]} \quad (8.1)$$

with

$$\text{pKa} = 0.96 + 0.90 \cdot \text{pH} - 0.039 \cdot \text{pH}^2 \quad (8.2)$$

where:

- DOC = the concentration of dissolved organic carbon (mg.l<sup>-1</sup>)
- m = the concentration of acidic functional groups on DOC (μmol<sub>c</sub>.mg<sup>-1</sup> C)
- Ka = the dissociation constant for organic acid (mol.l<sup>-1</sup>)
- [H] = the proton concentration (mol.l<sup>-1</sup>)

For m a value of 5.5 μmol<sub>c</sub>.mg<sup>-1</sup> C was used, based on Hendriksen and Seep (1980). Checks on the ionic balance were only made when all cations and anions were measured. The only allowances made were situations were (i) Al was missing at a pH > 5, (ii) alkalinity was missing

at a pH < 5 and (iii) DOC was missing (separate calculation). Checks on ionic balance could only be made for part of the measurements due to neglect of Na and Cl (for those ions only 843 measurements were available) and quite often also NH<sub>4</sub> and NO<sub>3</sub>. Actually, Na and Cl were optional parameters, but for a quality check it is absolutely necessary to include them. From a total number of 4971 measurements, checks on the complete ionic balance could be made at 312 measurements only. For another 643 measurements, checks could be made without including DOC, whereas no checks could be made at the remaining 4128 measurements. Compared with atmospheric deposition, graphs of the sum of cations versus the sum of anions showed less differences. (Fig 8.4).

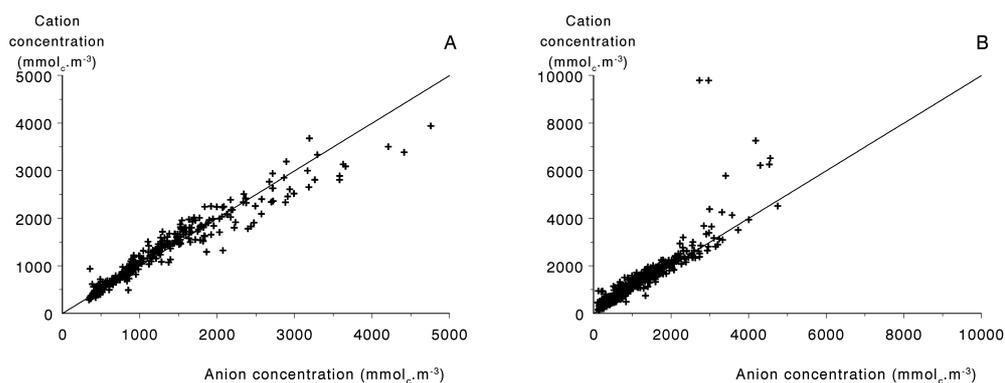


Figure 8.4 Relationships between the sum of cations and the sum of anions using measurements including DOC (A; 312 measurements) and excluding DOC (B; 643 measurements). The solid line represents a 1:1 line.

The percentage of measurements in an acceptable range of  $\pm 20\%$  was nearly 90% when DOC is taken into account, but it decreased to less than 60% when DOC was neglected (Table 8.6). In that case, there is generally a large cation excess. This result implies that a reliable charge balance check can only be made when all major cations and anions, including DOC, are available.

Table 8.6 Percentage of measurements in different ranges for the difference in the sum of cations (and the sum of anions) relative to the sum of cations

Difference (%)	Percentage	
	Complete ionic balance <sup>1)</sup>	Complete balance except DOC <sup>2)</sup>
< -30	1.0	0.3
-30 - -20	4.8	0.5
-20 - -10	11.9	3.6
-10 - 0	20.5	11.8
0 - 10	36.5	17.7
10 - 20	18.3	25.0
20 - 30	5.4	17.1
> 30	1.6	24.0

<sup>1)</sup> The total number of measurements equals 312

<sup>2)</sup> The total number of measurements equals 643

### Electric conductivity

A check on the difference between measured and calculated electric conductivity (EC) was only possible for 579 measurements, since the calculation of EC requires the availability of all major cations and anions (only DOC is not necessary). Furthermore, the number of EC measurements was limited, because EC is an optional parameter for the soil solution. Calculations were carried out according to Eq. 6.1, using the values for equivalent ionic conductivity given in Table 6.4. For Al, which is not included in Table 6.4 since this ion is mostly negligible in atmospheric

deposition, an equivalent conductance of  $0.06 \text{ kS.cm}^2.\text{eq}^{-1}$  was used. Results of the calculation gave a reasonable comparison with measured values (Fig. 8.5).

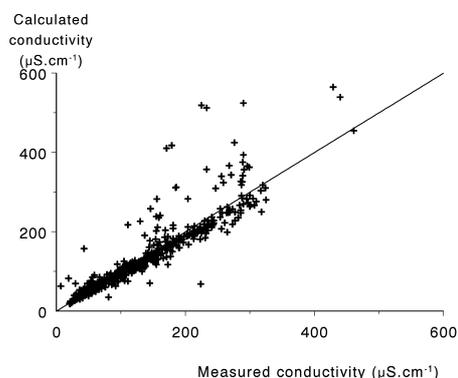


Figure 8.5 Relationship between the calculated and measured conductivity in soil solution using 579 individual measurements. The solid line represents a 1:1 line.

Considering an acceptable difference of 20% (WMO, 1992), approximately 85% of the measurements were within this criterion. Only 9.0% of the measurements differed by more than 30%. As with the difference between cations and anions, this result is better than those obtained for the atmospheric deposition data.

### ***Ionic ratios***

As with atmospheric deposition, the Na/Cl ratio in soil solution should somehow resemble those in sea water, since interactions of Na and Cl in the soil solution are small. Information on Na and Cl is, however, limited since both parameters are optional for the soil solution. Results of a regression between Na and Cl gave a slope of  $0.873 \text{ eq.eq}^{-1}$  with 55.3% of variance accounted for (fig. 8.6).

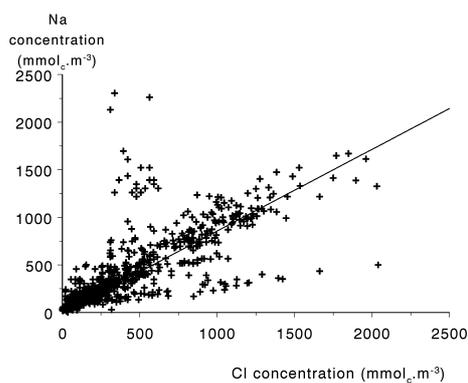


Figure 8.6 Relationships between the concentrations of Na and Cl in 843 individual measurements of soil solution chemistry. The solid line represents a 1:1 line.

The slope strongly resembles the Na/Cl ratio in sea water of  $0.858 \text{ eq.eq}^{-1}$ . As with atmospheric deposition, the Na/Cl ratios varied strongly. 90% of the values ranged between approximately 0.4 and 3.6. The high Na concentrations at relative low Cl concentrations (approximately  $500 \text{ mmol}_c.\text{m}^{-3}$ ; see Fig. 8.6) may be due to Na release from ordinary glass (sodium borosilicate) bottles, that are often used to collect samples in the field situation (Derome and Lindroos, 1997;

see also Annex 1) This is especially a problem with soil solution sampled by suction cups, because the samples are kept in those bottles in the field for up to two weeks.

Another relevant test is the relationship between Al and pH and alkalinity and pH. In general, Al should be negligible above pH 4.5 – 5.0, whereas the reverse is true for alkalinity. Results showed that this was mostly the case. There was only one outlier for Al at pH 5.5 (Fig 8.7A) and one outlier for alkalinity at pH 4.3 (Fig. 8.7B).

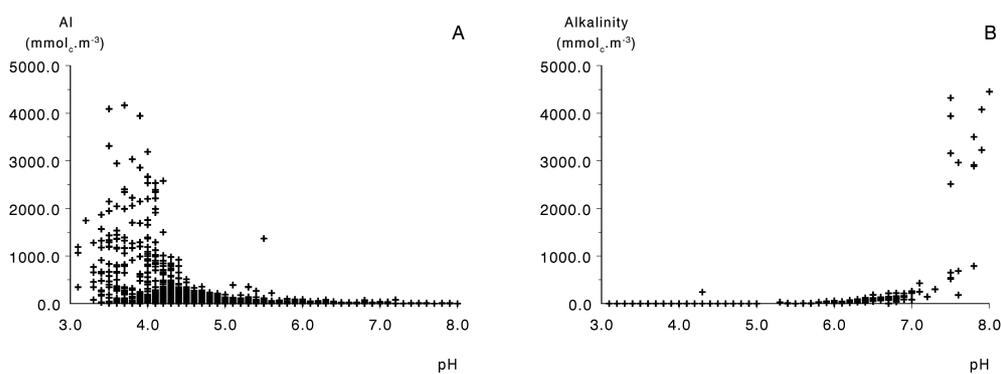


Figure 8.7 Relationships between the Al concentration and pH (A; 3637 measurements) and alkalinity and pH (B; 138 measurements).

## 8.2.4 Data evaluation

### *Assessment of relationships between ion concentrations in soil solution*

Relationships between the concentrations of Al, Ca, SO<sub>4</sub> and NO<sub>3</sub> and the pH, which are all key parameters with respect to the soil solution chemistry, were investigated by simple regression relationships. With respect to Al, the relation with SO<sub>4</sub> and NO<sub>3</sub> was investigated, while focusing on acid soils (pH < 4.5 or base saturation below 25%) where the acid input is assumed to be buffered by Al release mainly. Similarly, relationships between Ca and SO<sub>4</sub> and NO<sub>3</sub> were investigated, while focusing on slightly acid soils (pH > 4.5 or base saturation above 25-50 %) where the acid input is assumed to be buffered by Ca release mainly.

The relationship between Al concentration and pH was further investigated by relating the logarithmic free Al activity to the pH. Slopes of this relationship give some insight in the buffer mechanism of Al. In case of equilibrium release of an Al hydroxide (e.g. gibbsite) the slope should be 3. When Al release is due to interactions between protons and Al on soil organic matter, the slope may have a non-integer between 1 and 2 depending on the type of binding (Berggren, 1992, Wesselink et al., 1996). The activity of free (uncomplexed) Al was calculated with the equilibrium model Mineql (Schecher and Mc Avoy, 1994) by including complexation reactions with SO<sub>4</sub>, Cl, OH and extending the model with complexation by organic anions, which are most important with respect to Al. An important reason for calculating the activity of free (uncomplexed) Al is further that this form is most toxic to roots. Al complexed to organic anions hardly has a toxic impact. Polymeric Al can also be phytotoxic but it is debatable how common these polymeres are in nature. Inclusion of organic acid complexation reactions was only possible for plots where DOC data were available. The total concentration of so-called organic functional groups were related to the DOC by multiplying the DOC concentration with the concentration of acidic functional groups (5.5 mmol<sub>c</sub> per kg DOC). The dissociation of these functional groups and

the interaction with Al was described by using a so-called triprotic acid analog (Santore et al., 1996). The complexation constants used can also be found in this publication.

### ***Use of class limits to present results***

In order to evaluate the results of the soil solution chemistry in terms of possible negative impacts, use was made of critical chemical values for the concentrations of NO<sub>3</sub> and Al and the molar ratios of Al/(Ca+Mg+K) and NH<sub>4</sub>/Mg. Criteria thus used with an explanation of its background, are given in Table 8.7.

*Table 8.7 Critical chemical values for the concentrations of NO<sub>3</sub> and Al and the molar ratios of Al/(Ca+Mg+K) and NH<sub>4</sub>/Mg*

	Concentration (mmol <sub>e</sub> .m <sup>-3</sup> )		Molar ratio (mol.mol <sup>-1</sup> )	
	NO <sub>3</sub>	Al	NH <sub>4</sub> /Mg	Al/(Ca+Mg+K)
1 (low)	<100 <sup>1)</sup>	<20 <sup>2)</sup>	<1	<0.5
2 (intermediate)	100 - 400	20 - 200	1 - 5	0.5 - 1.0 <sup>5)</sup>
3 (high)	>400 <sup>2)</sup>	>200 <sup>3)</sup>	>5 <sup>4)</sup>	>2.5

<sup>1)</sup> Clearly elevated NO<sub>3</sub> concentration (Gunderssen et al., 1998) that may be related to vegetation changes (Warfinge et al., 1992)

<sup>2)</sup> Target value for ground water quality

<sup>3)</sup> Critical value related to effects on tree roots (Cronan et al., 1989)

<sup>4)</sup> Critical value related to decreased base cation uptake (Roelofs et al., 1985; Boxman et al., 1988)

<sup>5)</sup> Most common range of critical values related to adverse impacts on roots, such as root growth and root uptake, depending upon tree species (Sverdrup and Warfinge, 1993).

### ***Assessment of relationships between soil solution chemistry, stand and site characteristics, soil chemistry and atmospheric deposition***

Soil solution chemistry is not only influenced by the atmospheric input to and the chemical interactions in the soil, but also by the impacts of nutrient cycling, especially in the forest topsoil, and the water fluxes through the system. In order to investigate such relationships, use was made of both ordination techniques (Section 3.2) and multiple regression models (Section 3.3.2) relating responses or parameters (soil solution concentrations) to all predictor variables (stand and site characteristics and stress factors, such as atmospheric deposition and meteorological conditions) affecting this response. Information on hydrological fluxes was not available. Instead, use was made of information on meteorological data (measured precipitation and calculated precipitation excess) and of stand and site characteristics (soil type and tree species) which all affect this flux.

An overview of the predictor variables included in the various regression models is given in Table 8.8. With respect to atmospheric inputs, use was made of both throughfall and calculated total deposition data. Results are, however, limited to throughfall since calculations of total deposition are not so adequate on a plot basis (Section 6.3.1). This implies that the correlation between total deposition and soil solution chemistry will be negatively influenced by less reliable calculations. Furthermore, throughfall is the best estimate for the input to the soil solution. Whenever relevant, reference is made to correlations with calculated total deposition as well.

Precipitation data were taken from the deposition survey, since (more detailed) data from the meteorological survey were limited. Furthermore, on an annual basis precipitation data appeared to be comparable in both surveys (Section 7.2.3). The precipitation excess was calculated by subtracting potential evapotranspiration from the precipitation, using an external database. The

external database was used because the number of Intensive Monitoring plots with meteorological data, allowing the calculation of potential evapotranspiration was too limited (Section 7.2).

Table 8.8 Overview of the predictor variables used to explain the chemical composition of the soil solution at 53 Intensive Monitoring plots where both deposition data (throughfall) and soil solution chemistry data were available.

Predictor variables	SO <sub>4</sub>	NO <sub>3</sub>	NH <sub>4</sub>	N	Ca	Mg	K	Al	pH
<i>Site/Stand characteristics</i>									
Tree species	X	X	X	X	X	X	X	X	X
Soil type	X	X	X	X	X	X	X	X	X
<i>Throughfall</i>									
SO <sub>4</sub>	X								
NO <sub>3</sub>		X							
SO <sub>4</sub> +NO <sub>3</sub>					X	X	X	X	X
NH <sub>4</sub>		X	X		X	X	X	X	X
NH <sub>4</sub> +NO <sub>3</sub> (N)				X					
Ca					X				
Mg						X			
K							X		
BC* <sup>1)</sup>								X	X
<i>Meteorology</i>									
Precipitation	X	X	X	X	X	X	X	X	X
Prec. excess	X	X	X	X	X	X	X	X	X
<i>Soil chemistry</i>									
C/N ratio		X	X	X					
Base saturation					X	X	X	X	X
pH-CaCl <sub>2</sub>								X	

<sup>1)</sup> BC\* = Ca + Mg + K + Na - Cl

## 8.3 Results and discussion

### 8.3.1 Relationships between ion concentrations in soil solution

#### *Aluminium and strong acid anions*

The Al concentration in both topsoil and subsoil was strongly related to the concentration of SO<sub>4</sub> and NO<sub>3</sub> at a base saturation below 25% in the subsoil (Table 8.9). This implies that the acidic deposition is mainly neutralised by Al release in those acidic soils. This result is in line with model simulations (Reuss, 1983) and laboratory experiments (De Vries, 1994) indicating a significant Al release below a base saturation of 25%. Above a base saturation of 25%, there was no relationship between the concentration of Al versus SO<sub>4</sub> plus NO<sub>3</sub>, indicating that the acidity is mainly neutralised by the release of base cations (see below).

Table 8.9 Results of a linear regression between the Al concentration and the SO<sub>4</sub>+NO<sub>3</sub> concentration at different base saturation classes

Base saturation class (%)	Mineral topsoil				Mineral subsoil			
	N	$\alpha_0$	$\alpha_1$	$R^2_{adj}$	N	$\alpha_0$	$\alpha_1$	$R^2_{adj}$
< 25%	1954	45	0.35	68	133	-95	0.74	86
> 25%	801	65	-0.069	1	22	180	-0.0073	-
all	2755	46	0.34	65	155	250	0.30	29

A comparably sharp distinction was obtained by dividing the results in two pH ranges. As with base saturation, an increasingly strong relationship between Al and SO<sub>4</sub> plus NO<sub>3</sub> was observed going from the topsoil ( $R^2_{adj} = 68\%$ ) to the subsoil ( $R^2_{adj} = 84\%$ ) at a pH below 4.5 (Fig. 8.8).

Above pH 4.5, there was no relationship between the concentration of Al, SO<sub>4</sub> and NO<sub>3</sub> ( $R^2_{\text{adj}} = 3\%$  for both topsoil and subsoil).

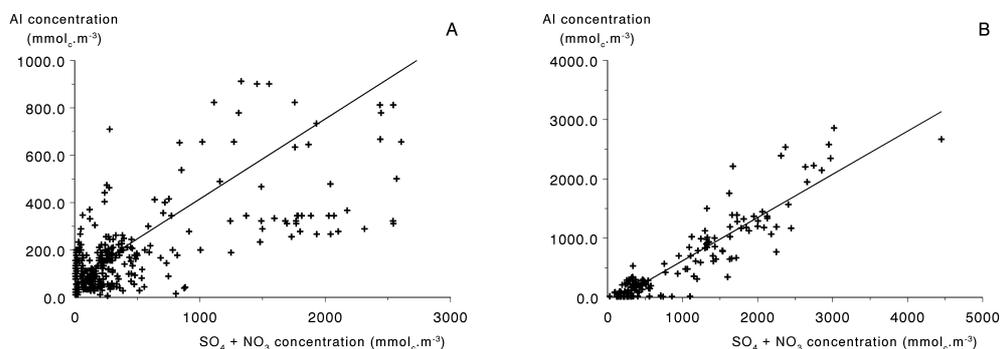


Figure 8.8 Relationship between the concentration of Al and the SO<sub>4</sub>+NO<sub>3</sub> concentration in the topsoil (A; 854 measurements) and subsoil (B; 159 measurements) of Intensive Monitoring plots with a pH < 4.5. The solid line represents a regression line.

### Calcium and strong acid anions

The relationship between Ca and strong acid anions was generally weaker in the topsoil than in the subsoil. Opposite to Al, the correlation mostly increased at plots with a high base saturation (Table 8.10). The results are limited to non-calcareous plots, where the influence of bicarbonate is relatively small.

Table 8.10 Results of a linear regression between the Ca concentration and the SO<sub>4</sub>+NO<sub>3</sub> concentration at different base saturation classes in non-calcareous soil.

Base saturation class (%)	Mineral topsoil				Mineral subsoil			
	N	$\alpha_0$	$\alpha_1$	$R^2_{\text{adj}}$	N	$\alpha_0$	$\alpha_1$	$R^2_{\text{adj}}$
< 25%	1961	41	0.13	37	135	8.8	0.16	78
>25	827	-12	0.97	80	22	-130	0.86	97
All	2788	42	0.14	33	157	-300	0.58	66

The difference in relationships between the concentration of Ca and SO<sub>4</sub> plus NO<sub>3</sub> at low and high base saturation is further illustrated in Fig. 8.9.

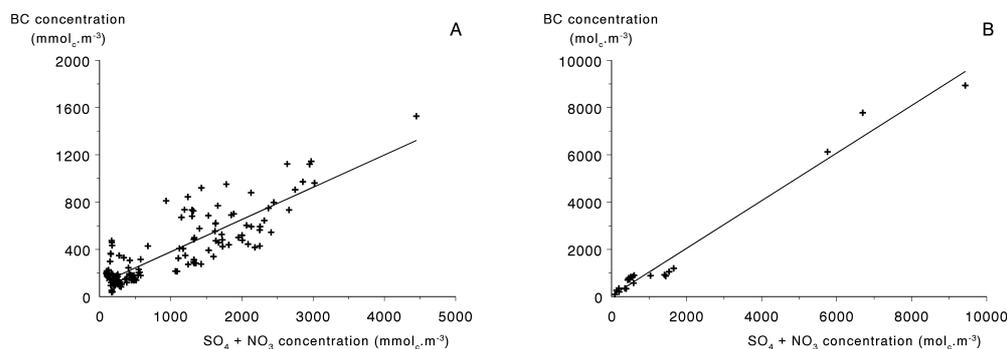


Figure 8.9 Relationship between the concentration of BC and the SO<sub>4</sub>+NO<sub>3</sub> concentration in the subsoil of Intensive Monitoring plots with a base saturation below 25% (A; 135 measurements) and above 25% (B; 22 measurements). The solid line represents a regression line.

The figure shows that (i) the correlation increases with an increased in base saturation ( $R^2_{\text{adj}} = 78\%$  at a base saturation below 25% and 99% at a base saturation above 50%) and (ii) the slope

much more strongly resembles the 1 : 1 line at high base saturation (see also the slopes in Table 8.10).

### Aluminium and pH

The logarithmic concentration of Al was clearly related to the pH, when using all available data for the topsoil (Fig. 8.10A) and subsoil (Fig. 8.10B). However, the relationship was not very strong, especially in the topsoil since the occurrence of complexation strongly influences the total Al concentration without having a strong effect on pH ( $R^2_{\text{adj}} = 56\%$  for the topsoil and 60% for the subsoil).

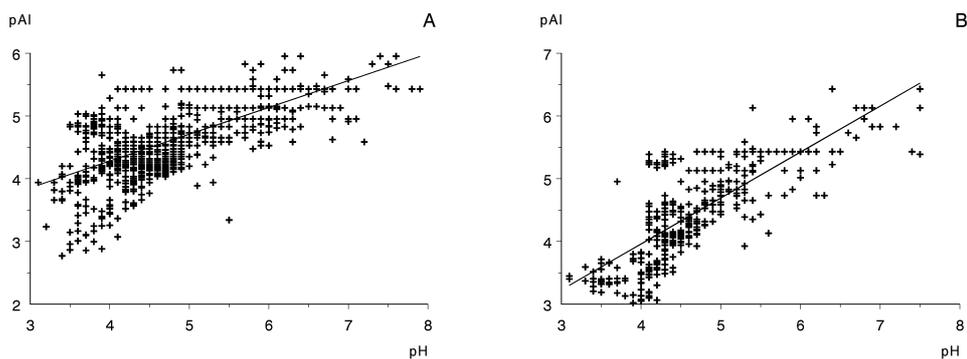


Figure 8.10 Relationships between the pAl (the negative logarithmic total concentration of Al) and the pH in the mineral topsoil (A; 2810 measurements) and subsoil (B; 409 measurements) of Intensive Monitoring plots. The solid line represents a regression line.

A much better relationship with pH is to be expected with the concentration (or even better, the activity) of free Al. Free Al could, however, only be derived for a very limited number of measurements in the mineral soil, since DOC which is crucial to calculate the free Al activity because of the dominating influence of organic Al complexes (Section 8.2.4), was hardly measured there (Section 8.2.3). Information of DOC was, however, largely available for the organic layer. Results of the relationship between the calculated logarithmic free Al concentration and pH in this layer, indeed, appeared to be much stronger ( $R^2_{\text{adj}} = 74\%$ ) than between the logarithmic total Al concentration and pH ( $R^2_{\text{adj}} = 20\%$ ) (Fig. 8.11).

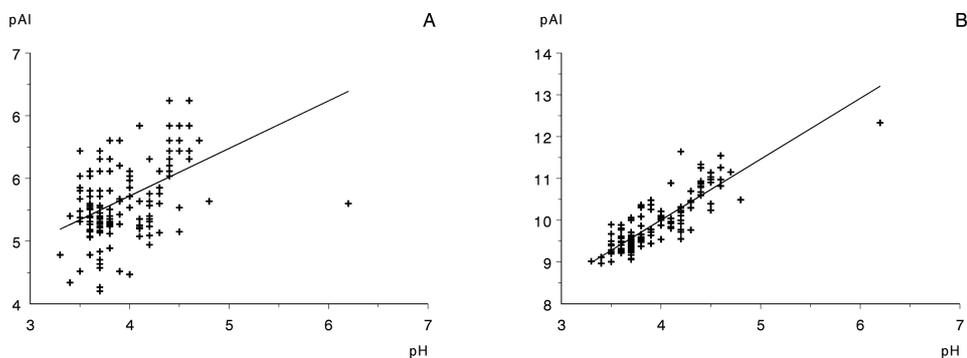


Figure 8.11 Relationships between the pH and the (negative) logarithmic total Al concentration (A; 149 measurements) and the free Al activity (B; 149 measurements) in the organic layer of 14 Intensive Monitoring plots in the Netherlands and Ireland. The solid line represents a regression line.

The slopes appeared to increase from 0.57 using total Al concentrations to 1.39 using free Al activities. A non-integer value of the slope between 1 and 2 implies that Al release in the organic layer is most likely dominated by equilibrium complexation reactions with soil organic matter.

### 8.3.2 Ranges in ion concentrations and ion ratios in view of critical levels.

Information on ion concentrations and ion ratios is given below for the organic layer, mineral topsoil (0-40 cm) and mineral subsoil (40-80 cm) at a total of 103 Intensive Monitoring plots. Values refer to annual average concentrations, which were also used in the statistical analyses with environmental factors, such as stand and site characteristics, atmospheric deposition and meteorological factors (Section 8.3.3). Another reason for using annual average data is to avoid bias of plots with a much higher frequency of measurements in time. The number of plots depends on the layer considered since not all layers are included at all plots (see Table 8.1). Furthermore, several ions, such as NO<sub>3</sub> and NH<sub>4</sub> were measured at less plots. The number of plot-layer combinations that were used for the various ions are given in Table 8.11. In comparing frequency distributions of different ions, those differences in data availability should be kept in mind.

Table 8.11 Numbers of plot/layer combinations that were used to describe the concentration ranges for the major ions in soil solution

Soil layer	Number <sup>1)</sup>											
	pH	Al	Ca	Mg	K	Na	NH <sub>4</sub>	SO <sub>4</sub>	NO <sub>3</sub>	Cl	Alk	DOC
Organic layer	21	21	21	21	21	15	15	21	21	15	0	21
Topsoil (0-40 cm)	88	83	87	87	87	47	47	87	84	47	12	61
Subsoil (40-80 cm)	115	112	116	116	116	48	48	116	89	48	9	90

<sup>1)</sup> Note that the number can be larger than the number of plots since several layers may be sampled in both the topsoil and subsoil.

#### ***Ion concentrations***

Concentrations of SO<sub>4</sub>, NO<sub>3</sub>, and total N were mostly lower than 2000 mmol<sub>c</sub>.m<sup>-3</sup>, whereas concentrations of NH<sub>4</sub> were nearly always lower than 1000 mmol<sub>c</sub>.m<sup>-3</sup> (Fig 8.12). Concentrations of SO<sub>4</sub> and NO<sub>3</sub> seem to decrease going from the organic layer to the mineral soil but this effect is influenced by the different number of measurements in the various layers (Fig 8.12A,B). In general, concentrations increase from the organic layer to the mineral layer, partly due to a decrease in water flux in this direction. Differences between the mineral topsoil and the mineral subsoil were less significant. As expected, NH<sub>4</sub> clearly decreased going from the organic layer to the mineral topsoil and the mineral subsoil (Fig 8.12C). Elevated NH<sub>4</sub> concentrations in the organic layer are most likely due to the occurrence of mineralisation in this layer. Compared to the relatively constant N concentration with depth (Fig 8.12D), the decrease in NH<sub>4</sub> concentration indicates the occurrence of preferential NH<sub>4</sub> uptake and/or nitrification with depth.

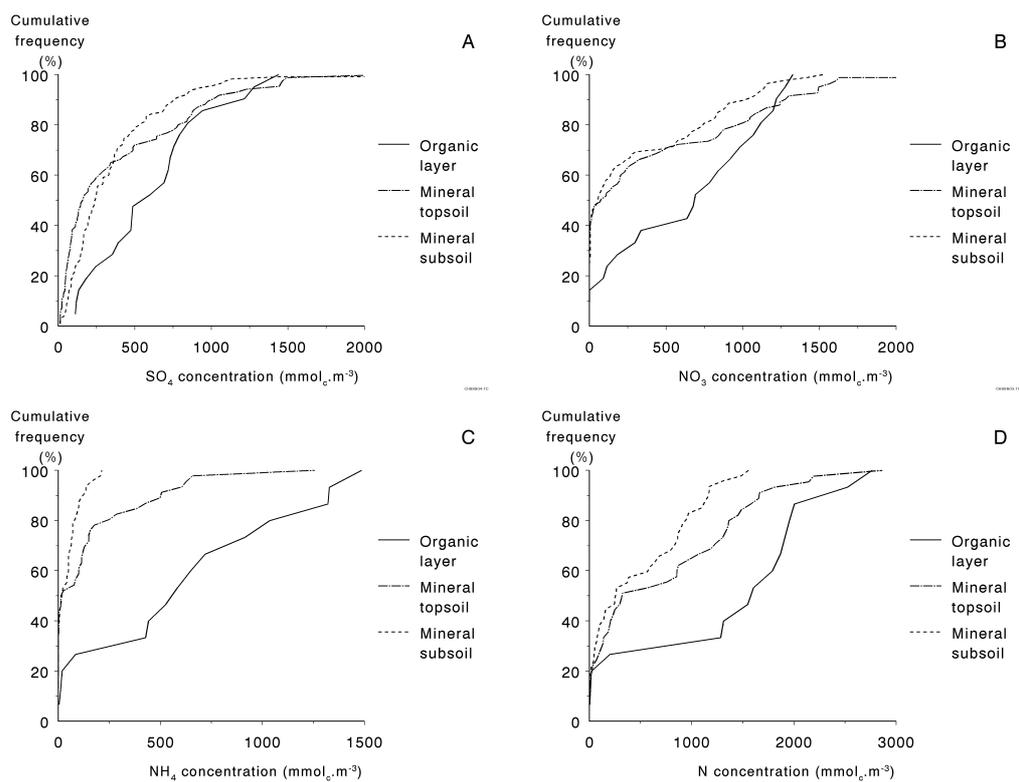


Figure 8.12 Cumulative frequency distributions of the concentrations SO<sub>4</sub> (A), NO<sub>3</sub> (B), NH<sub>4</sub> (C) and total N (D) in the organic layer, mineral topsoil (0-40 cm) and mineral subsoil (40-80 cm) of 103 Intensive Monitoring plots

Concentrations of the major cations Al and Ca in the mineral soil occurred in a similar range as SO<sub>4</sub> and NO<sub>3</sub> (Fig 8.13A, B), already indicating a correlation between those ions (see Section 8.3.2). Opposite to SO<sub>4</sub>, NO<sub>3</sub> and NH<sub>4</sub>, the concentration of Al increased significantly going from the organic layer to the mineral topsoil and the mineral subsoil. Differences were largest between the organic layer and the mineral soil. This is to be expected, since the major source of Al is the dissolution from amorphous compounds in the mineral soil followed by complexation on organic matter.

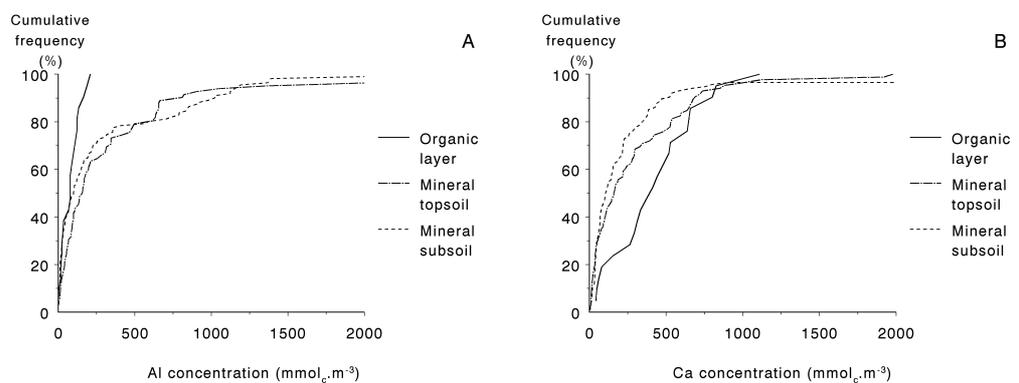


Figure 8.13 Cumulative frequency distributions of the concentrations of Al (A) and Ca (B), in the organic layer, mineral topsoil (0-40 cm) and mineral subsoil (40-80 cm) of 103 Intensive Monitoring plots

The concentrations of NO<sub>3</sub> in the subsoil exceeded the official ground water quality standard of 800 mmol<sub>c</sub>·m<sup>-3</sup> at 14% of the plots. The ground water quality standard of 20 mmol<sub>c</sub>·m<sup>-3</sup> for Al was

exceeded at more than 70% of the plots at greater depth. An Al concentration of  $200 \text{ mmol}_c.\text{m}^{-3}$ , that has sometimes be considered indicative for negative impacts on tree roots, was exceeded at approximately 40% of the plots (Table 8.12). Note, however, that the concentration of the uncomplexed (free) aluminium, which is considered toxic to roots is much lower. This implies that the actual number of plots exceeding a critical value is probably lower as well.

Table 8.12 The percentage of observations of  $\text{NO}_3$  and Al concentrations in the mineral topsoil and subsoil between different class limits

$\text{NO}_3$			Al		
concentration class ( $\text{mmol}_c.\text{m}^{-3}$ )	%topsoil (n=84)	%subsoil (n=89)	concentration class ( $\text{mmol}_c.\text{m}^{-3}$ )	%topsoil (n=83)	%subsoil (n=112)
< 100	51	56	< 20	11	24
100 – 400	17	13	20 – 200	50	42
> 400	32 <sup>1)</sup>	30 <sup>1)</sup>	> 200	39	34

<sup>1)</sup> The percentage of observations above the EC drinking water quality standard of  $800 \text{ mmol}_c.\text{m}^{-3}$  ( $50 \text{ mg.l}^{-1}$ ) was 26% in the topsoil and 18% in the subsoil

The pH, which is significantly related to the Al concentration and the free Al ion activity (see Section 8.3.2) varied between 3 and 8.5 in all layers. The pH values, however, significantly increased with depth, indicating the occurrence of buffering reactions (release of base cations and Al by weathering and cation exchange) in this direction (Fig 8.14A). Apart from pH, dissolved organic carbon (DOC) also has a profound influence on the concentration (activity) of the free Al ion, which is most toxic to roots. This is due to complexation of Al with DOC. DOC concentrations mostly decreased from the organic layer to the mineral topsoil, whereas a large decrease took place with depth in the mineral soil. Concentrations of DOC generally ranged between 20 and  $150 \text{ mg.l}^{-1}$  in the organic layer and the mineral topsoil and between 10 and  $50 \text{ mg.l}^{-1}$  in the mineral subsoil (Fig 8.14B).

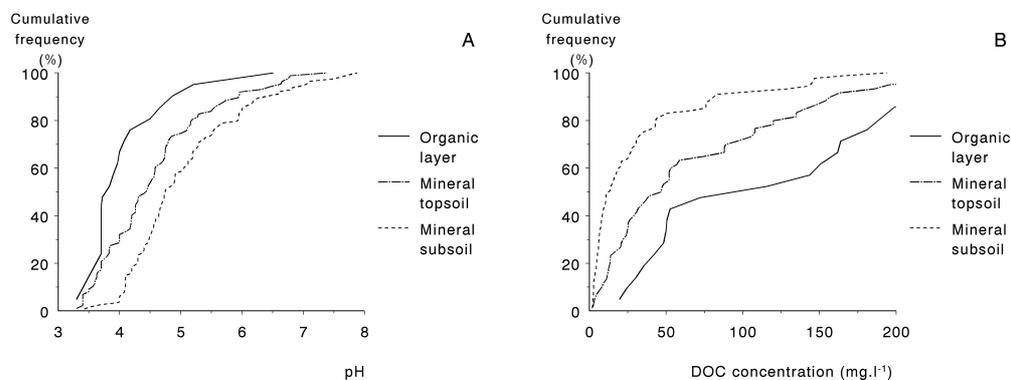


Figure 8.14 Cumulative frequency distributions of the pH (A) and the DOC concentration (B) in the organic layer, mineral topsoil (0-40 cm) and mineral subsoil (40-80 cm) of 103 Intensive Monitoring plots

### Ion ratios

As with Al concentrations, the ratios of Al to Ca or the sum of Ca, Mg and K increased with depth. Al/Ca ratios were generally less than 0.5 in the organic layer and less than 5.0 in the mineral soil (Fig. 8.15A), whereas Al/(Ca+Mg+K) ratios were mostly less than 2.0 in the mineral soil (Fig. 8.15B). Opposite to Al/base cation ratios, the ratios of  $\text{NH}_4$  to base cations decreased with depth. Both the  $\text{NH}_4/\text{K}$  ratio and  $\text{NH}_4/\text{Mg}$  ratio were mostly less than 1.0 in the mineral soil, whereas values up to 5.0 were frequently observed in the organic layer (Fig. 8.15C, D).

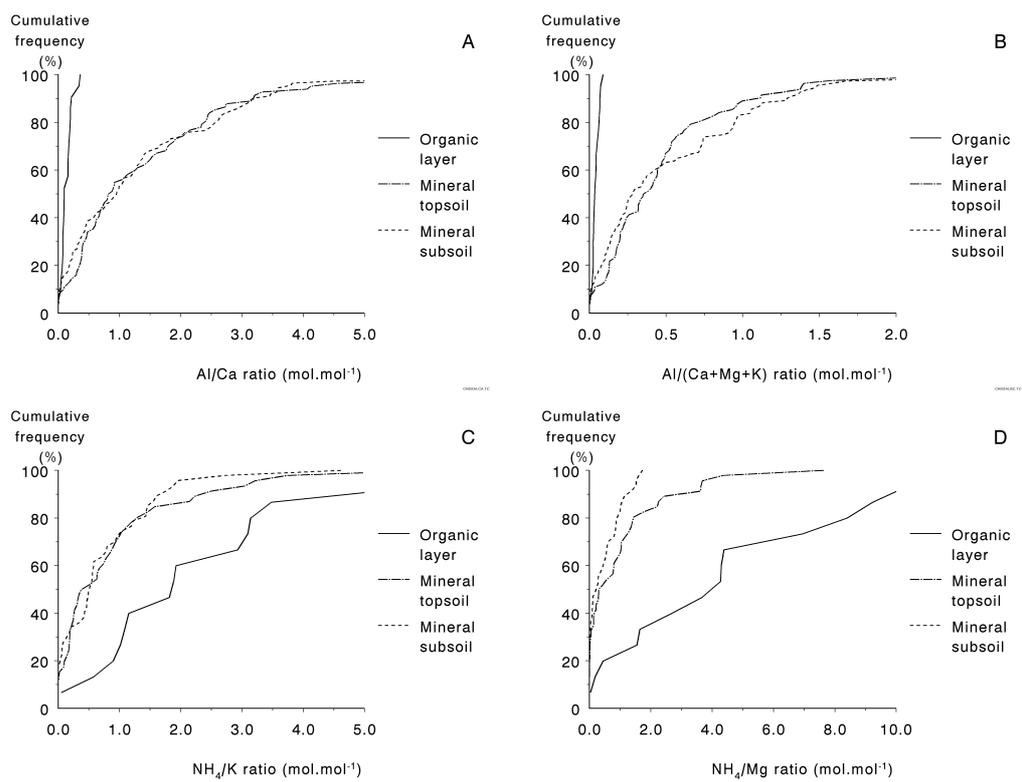


Figure 8.15 Cumulative frequency distributions of the ratios of Al/Ca (A), Al/(Ca+Mg+K) (B), NH<sub>4</sub>/K (C) and NH<sub>4</sub>/Mg (D) in the organic layer, mineral topsoil (0-40 cm) and mineral subsoil (40-80 cm) of 103 Intensive Monitoring plots

The number of plots exceeding a critical Al/(Ca+Mg+K) ratio was approximately 10-15%, depending upon the soil layer considered (Table 8.13). Using a critical Al/Ca ratio of 1.0, as suggested by Ulrich and Matzner (1983), this number increased to approximately 40-45%. Note, however, that critical ratios refer specifically to the concentration of free (uncomplexed) Al, that is toxic to roots, whereas the calculated ratios refer to total Al. This is because free Al could not be calculated in most plots, due to missing DOC concentrations. Furthermore, critical ratios do depend on the tree species considered (Sverdrup and Warfinge, 1993). Finally, in the uppermost soil layer (0-20 cm), where most nutrient uptake occurs, Al to base cation ratios are mostly lower. Consequently, those percentages should be seen as worst case estimates. Critical NH<sub>4</sub>/Mg and NH<sub>4</sub>/K ratios of 5.0 were hardly ever exceeded in the mineral soil (see also 8.13).

Table 8.13 The percentage of observations of Al/(Ca+Mg+K) and NH<sub>4</sub>/Mg ratios in the mineral topsoil and subsoil between different class limits

Al/(Ca+Mg+K)			NH <sub>4</sub> /Mg		
ratio class (eq.eq <sup>-1</sup> )	%topsoil (n=83)	%subsoil (n=112)	ratio class (eq.eq <sup>-1</sup> )	%topsoil (n=47)	%subsoil (n=48)
< 0.5	67	63	< 1	64	79
0.5 – 1.0	22	20	1 – 5	34	21
> 1.0 <sup>1)</sup>	11	17	> 5 <sup>2)</sup>	2	0

<sup>1)</sup> For the Al/Ca ratio, the percentage of observations exceeding a critical value of 1.0 equalled 45% in the topsoil and 46% in the subsoil

<sup>2)</sup> The percentage of observations exceeding a critical NH<sub>4</sub>/K ratio of 5.0 was also nearly negligible (4% in the topsoil and 2% in the subsoil)

### 8.3.3 Influence of stand and site characteristics, atmospheric deposition, meteorology and soil chemistry on soil solution chemistry

#### 8.3.3.1 Principal component analyses

##### *Relationships between soil solution chemistry in the mineral topsoil and environmental factors*

Figure 8.16 shows the biplot resulting from the PCA analysis on a soil solution chemistry data set for the mineral topsoil (0 – 40 cm) in combination with both major site characteristics (deposition region, tree species and soil type), precipitation and major soil characteristics (base saturation and C/N ratio of both the organic layer and the mineral topsoil) for a total of 27 sites only. The first and second axis explain 77 and 10% of the total variance respectively. The predictor variables explain 78% of the variation in soil solution chemistry between sites, of which 94% is displayed in the diagram.

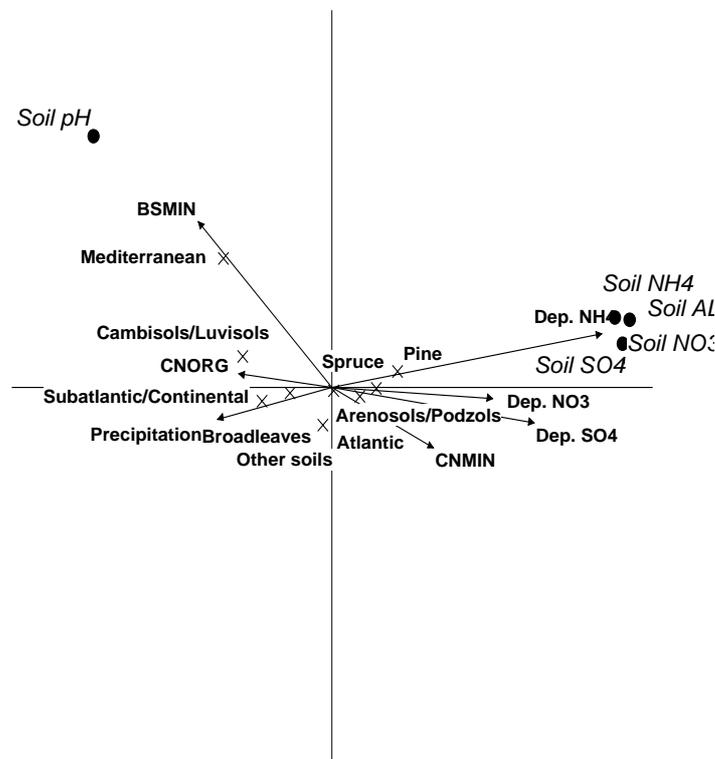


Figure 8.16 PCA diagram of the soil solution chemistry (response variables) in the mineral topsoil (0-40cm) in combination site and stand characteristics, precipitation, atmospheric deposition and soil chemistry (predictor variables). For interpretation of the diagram we refer to Section 3.3.2

The diagram shows that all soil solution chemistry parameters are correlated with each other,  $\text{SO}_4$ ,  $\text{NH}_4$  and  $\text{NO}_3$  and Al positively and pH negatively. The sulphate and nitrate concentrations are positively correlated with deposition data and negatively correlated with precipitation, most likely as a result of dilution.

##### *Relationships between soil solution chemistry in the mineral subsoil and environmental factors*

Figure 8.17 shows the biplot resulting from the PCA analysis on a soil solution chemistry data set for the mineral topsoil (40 – 80 cm) in combination with both major site characteristics

(deposition region, tree species and soil type), precipitation and major soil characteristics (base saturation and C/N ratio of the mineral subsoil) for a total of 18 sites only. The first and second axis explain 44 and 26% of the total variance respectively. The predictor variables explain 75% of the variation in deposition between sites, of which 76% is displayed in the diagram.

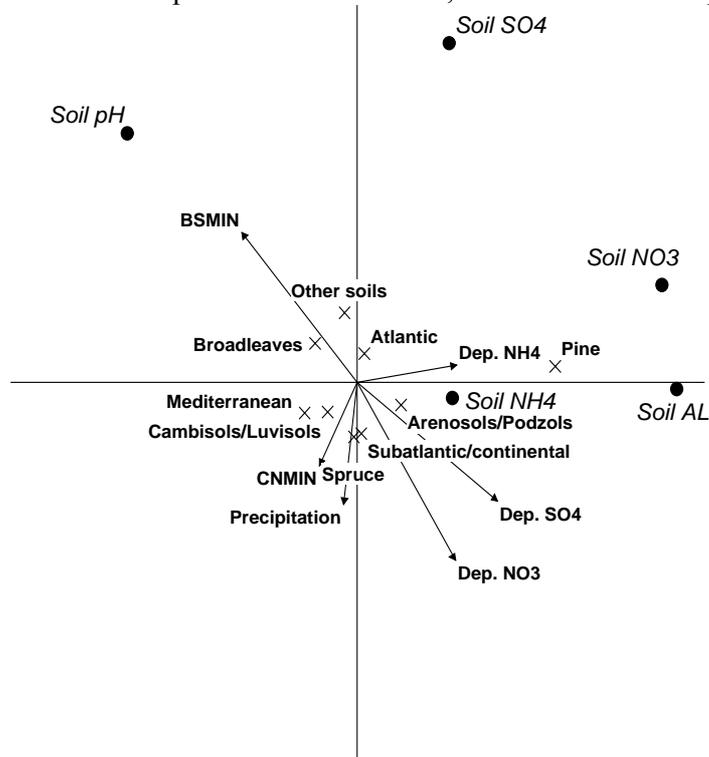


Figure 8.17 PCA diagram of the soil solution chemistry (response variables) in the mineral subsoil (40 – 80 cm) in combination site and stand characteristics, precipitation, atmospheric deposition and soil chemistry (predictor variables). For interpretation of the diagram we refer to Section 3.3.2

The diagram shows a weaker correlation between soil chemistry parameters compared to the 0 – 40 cm data set.

### 8.3.3.2 Multiple regression analyses

As with the results of the PCA, tree species and soil type never appeared to have any significant influence on the concentrations of the considered ions in both topsoil and subsoil. Results presented are thus limited to relationships with atmospheric deposition (throughfall), meteorology (precipitation or precipitation excess) and soil chemical data (C/N ratio, base saturation and/or pH-CaCl<sub>2</sub>) whenever relevant.

#### *Sulphur and nitrogen compounds*

A summarising overview of the results of multiple linear regression, using both the original and the log-transformed data for the concentration and deposition (throughfall) of S and N compounds, is given in Table 8.14. Results show that a logarithmic transformation of concentration data, deposition data and meteorological data significantly improved the relationships between environmental factors and the concentrations of SO<sub>4</sub> and NO<sub>3</sub>, but not for NH<sub>4</sub> and total N. The number of plots on which the results are based varied between 45 and 59 for

SO<sub>4</sub> and NO<sub>3</sub> and between 32-34 for NH<sub>4</sub> and total N. The limited number for the latter compounds is due to the fact that several countries do not measure NH<sub>4</sub>.

Table 8.14 Overview of the predictor variables explaining the concentrations of SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub> and total N in the topsoil (0-40 cm) and subsoil (40-80 cm) at 32-59 Intensive Monitoring plots, with the percentage variance accounted for<sup>1)</sup>.

Predictor variables	SO <sub>4</sub>		NO <sub>3</sub>		NH <sub>4</sub>		N	
	topsoil	subsoil	topsoil	subsoil	topsoil	subsoil	topsoil	subsoil
<i>Throughfall</i>								
SO <sub>4</sub>	++	++						
NO <sub>3</sub>			0	0				
NH <sub>4</sub>			++	++	++	++		
N							++	++
<i>Meteorology</i>								
Precipitation	--	0	0	0	- (0)	-	0 (--)	- (0)
Precipitation excess	0	- (--)	0 (--)	0	0 (+)	0	- (0)	
<i>Soil chemistry</i>								
C/N ratio			0 (-)	0	+	0	0	0
N <sup>2)</sup>	53	59	50	45	33	34	32	34
R <sup>2</sup> <sub>adj</sub> (%) <sup>3)</sup>	62 (84)	23 (54)	44 (82)	66 (61)	76 (79)	60 (58)	60 (55)	63 (59)

<sup>1)</sup> 0 = insignificant at the 95% level: t value < 2.0

+/- = significant: t value > 2.0

++/-- = highly significant: t value > 3.0

A '+' sign implies that the response variable (the concentration of SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub> or total N) increases with an increase in the predictor variable, whereas a '-' sign implies the opposite. Signs in brackets are related to results in which the logarithmic concentrations were used against the logarithm of the deposition and precipitation (excess).

<sup>2)</sup> N = number of plots.

<sup>3)</sup> R<sup>2</sup><sub>adj</sub> = percentage variance accounted for.

Using the original data, the explained variation (R<sup>2</sup><sub>adj</sub>) by environmental factors mostly ranged between 45 and 75%, with the exception of SO<sub>4</sub> in the subsoil where the explanation was only 23%. Using log-transformed data, the percentage of variance accounted for varied between 54 and 84%, the extremes being the result for SO<sub>4</sub> in the subsoil and the topsoil, respectively (Table 8.13). For SO<sub>4</sub> in both topsoil and subsoil and NO<sub>3</sub> in the topsoil, R<sup>2</sup><sub>adj</sub> increased by 20-40% using log-transformed concentration and deposition data instead of original data. For NO<sub>3</sub> in the subsoil and the concentrations of NH<sub>4</sub> and N, a logarithmic transformation did not improve the results.

In all situations, atmospheric deposition of the considered compound was the most important influencing factor with the exception of NO<sub>3</sub>. For this element, NO<sub>3</sub> deposition had no significant impact, not in the topsoil nor in the subsoil, whereas NH<sub>4</sub> deposition was highly significant (t value > 3.0) in both cases. The variation explained by atmospheric deposition data alone varied between 44% and 71% (with the exception of SO<sub>4</sub> in the subsoil; 19%) when using the original data. A slight increase in R<sup>2</sup><sub>adj</sub> was generally obtained by the inclusion of either the measured precipitation or a calculated precipitation excess.

A logarithmic transformation most strongly improved the results for NO<sub>3</sub> in the topsoil (R<sup>2</sup><sub>adj</sub> increased from 44% to 72%) as illustrated in Fig. 8.18. This is, however, mainly due to two outliers (High NO<sub>3</sub> concentrations) at an NH<sub>4</sub> deposition below 1000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>.

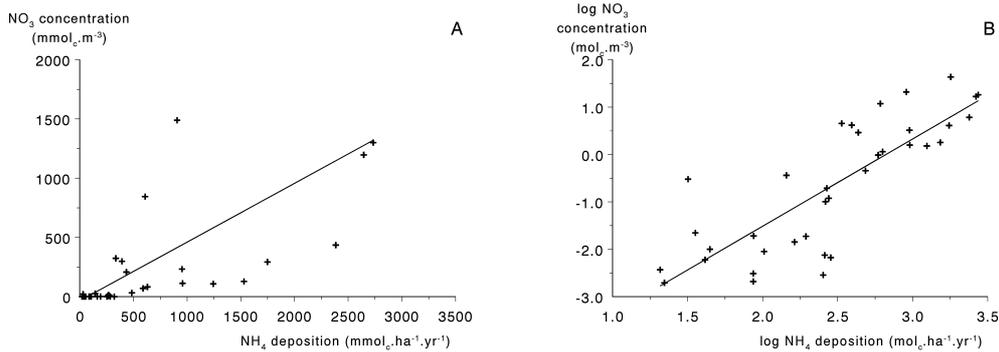


Figure 8.18 Relationships between the  $\text{NO}_3$  concentration in the topsoil and the  $\text{NH}_4$  deposition at .. Intensive Monitoring plots using original data (A) and log-transformed data (B). The solid line represents a regression line.

The highly significant impact of  $\text{NH}_4$  deposition on the concentration of both  $\text{NO}_3$  and  $\text{NH}_4$  in the topsoil and subsoil is further illustrated in Fig. 8.19. With the exception of  $\text{NO}_3$  in the topsoil (see Fig. 8.18B), results appeared to be better when using the original (not-transformed) that a simple linear regression is not a very appropriate description.

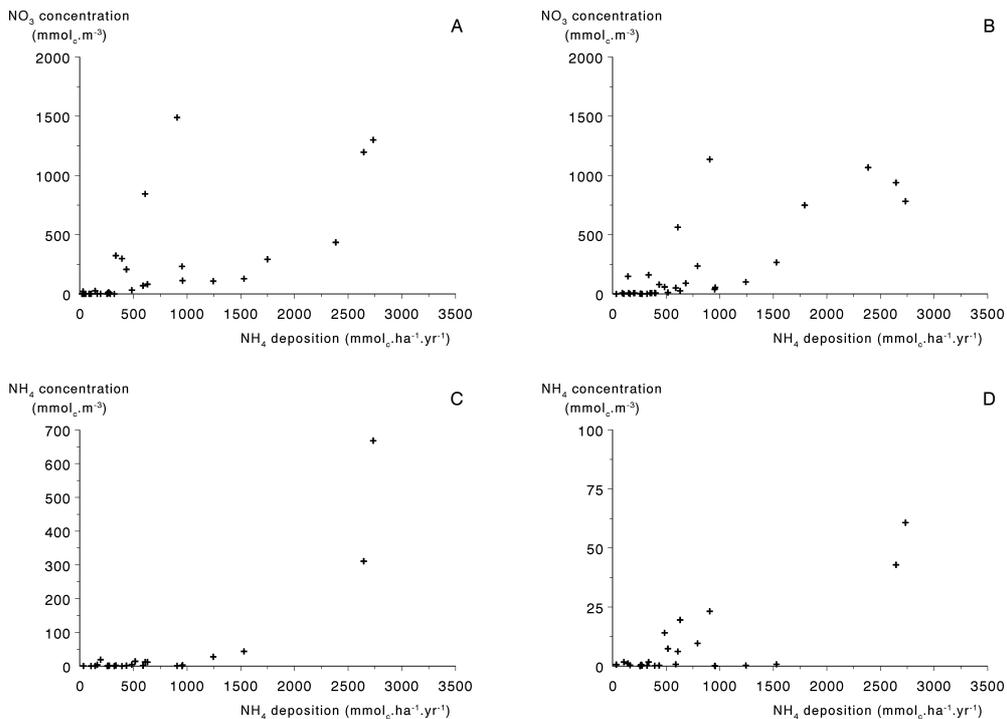


Figure 8.19 Relationships between the  $\text{NH}_4$  deposition and the  $\text{NO}_3$  and  $\text{NH}_4$  concentration in the topsoil (A, C) and subsoil (B, D) at 33-50 Intensive Monitoring plots.

In general, one can say that  $\text{NO}_3$  and  $\text{NH}_4$  concentrations are low at an  $\text{NH}_4$  deposition below 500 mmol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. Above this deposition level, concentrations generally increased with an increase in deposition level, but large variations did occur. Unlike results presented by Dise et al. (1998) and Gundersen et al. (1998), the influence of the C/N ratio on the measured  $\text{NO}_3$  and  $\text{NH}_4$  concentration was mostly negligible. In case of  $\text{NH}_4$  in the subsoil there was a significant effect, but this is likely to be an artefact since the 'sign' is opposite to what is expected (a '+' sign implies that the  $\text{NH}_4$  concentration increases with an increase in the C/N ratio). The small to insignificant influence of the C/N ratio on  $\text{NO}_3$  and  $\text{NH}_4$  concentrations was also observed in 150

forested stands in the Netherlands (De Vries and Leeters, 1999). A significant relationship was only found in a subset of well-drained soils with coniferous stands. A more in-depth analysis, based on much more data is necessary to further investigate the possible role of the C/N ratio of the soil on the N dynamics.

### **Base cations, aluminium and pH**

Results of the multiple regression analyses, using both the original and log-transformed data for the concentration of base cations and Al and the deposition of base cations, S and N compounds are presented in Table 8.15.

*Table 8.15 Overview of the predictor variables explaining the concentrations of SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub> and total N in the topsoil (0-40 cm) and subsoil (40-80 cm) at 32-50 Intensive Monitoring plots, with the percentage variance accounted for<sup>1)</sup>.*

Predictor variables	Ca		Mg		K		Al		pH				
	top.	sub.	top.	sub.	top.	sub.	top.	sub.	top.	sub.			
<i>Throughfall</i>													
Ca	++ (0)	++											
Mg				+	(++)								
K					+	(0)	-						
BC* <sup>2)</sup>							-	0	0				
SO <sub>4</sub> +NO <sub>3</sub>	0	--	0	0	--	(0)	0	(++)	0	--			
NH <sub>4</sub>	++	++ (+)	++	+	++	++ (0)	++	++ (0)	--	--			
<i>Meteorology</i>													
Precipitation	--	-	-	(--)	0	(-)	0	(--)	--	(-)	0	+	
Precipitation excess	0	0	0	-	(0)	0	0	0	0	0	0	0	
<i>Soil chemistry</i>													
Base saturation	0	(++)	0	(+)	0	(+)	+	0	0	0	0	++	++
pH-CaCl <sub>2</sub>								0	(--)	--			
N <sup>3)</sup>	50	44	50	45	50	45	47	42	51	44			
R <sup>2</sup> <sub>adj</sub> (%) <sup>4)</sup>	73 (59)	62 (32)	51 (54)	36 (60)	64 (48)	40 (14)	57 (77)	67 (83)	49	57			

<sup>1)</sup> 0 = insignificant at the 95% level: t value < 2.0

+/- = significant: t value > 2.0

++/-- = highly significant: t value > 3.0

A '+' sign implies that the response variable (the concentration of Ca, Mg, K, Al or pH) increases with an increase in the predictor variable, whereas a '-' sign implies the opposite. Signs in brackets are related to results in which the logarithmic concentrations were used against the logarithm of the deposition and precipitation (excess).

<sup>2)</sup> BC\* = Ca + Mg + K + Na - Cl

<sup>3)</sup> N = number of plots.

<sup>4)</sup> R<sup>2</sup><sub>adj</sub> = percentage variance accounted for.

With the exception of Al, the logarithmic transformation decreased the explained variation of cation concentrations by environmental factors. Results showed that the deposition of NH<sub>4</sub> had a (highly) significant impact on all the considered compounds, increasing the concentration of base cations and Al and decreasing the pH. This can be explained by the acidifying impact of NH<sub>4</sub> deposition, caused by the conversion of NH<sub>4</sub> to NO<sub>3</sub> (nitrification) in the soil. Unexpectedly, the deposition of SO<sub>4</sub>+NO<sub>3</sub> did not seem to have a significant impact on the base cation concentrations (Table 8.15). This result should, however, be interpreted with care. In general, there is a high correlation between the deposition of NH<sub>4</sub>, NO<sub>3</sub> and SO<sub>4</sub>. This implies that those predictor variables are partly exchangeable in a multiple regression model. Results of the analyses with one predictor variable only, showed that SO<sub>4</sub>+NO<sub>3</sub> was always a highly significant predictor for Ca, Mg and K, except for Ca in the subsoil. The impact of NH<sub>4</sub> deposition, however, appeared

to be more significant and consequently  $\text{SO}_4+\text{NO}_3$  generally appeared insignificant in a multiple regression model.

The influence of base cation deposition on base cation concentrations appeared to differ. For Ca it was highly significant in both topsoil and subsoil and for Mg in the subsoil only. For K and Mg in the topsoil, the impact was small to insignificant (Table 8.15). In general,  $\text{NH}_4$  deposition was an equally good predictor for the cation concentration as the throughfall of the cation itself. This is illustrated in Fig. 8.20 for K in the topsoil.

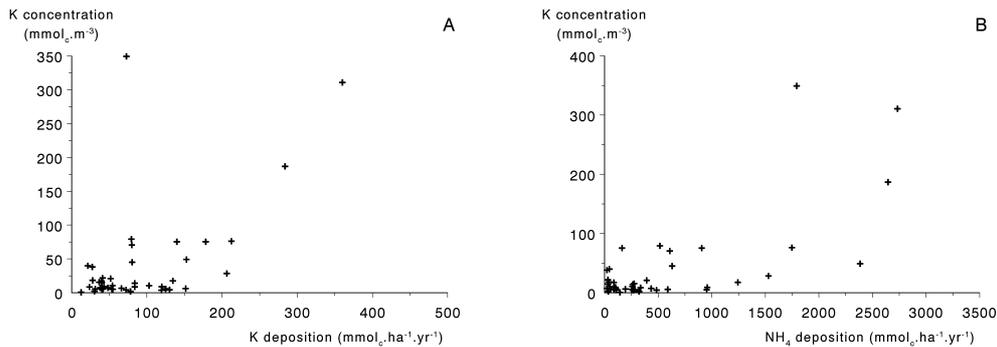


Figure 8.20 Relationship between the K concentration in the topsoil and the K deposition (A) and  $\text{NH}_4$  deposition (B) at 45-50 Intensive Monitoring plots.

Either the precipitation or the precipitation excess mostly was significant to highly significant, especially when using the log-transformed data. Base saturation did not have a significant influence on the base cation concentrations when using the original data, with the exception of Mg in the subsoil. Using log-transformed data, however, there was a significant impact on Ca and Mg in both topsoil and subsoil (Table 8.15). The insignificant impact on K is to be expected, since the occupation of K on the exchange complex is very limited.

Apart from the deposition of  $\text{NH}_4$  and for  $\text{SO}_4+\text{NO}_3$ , the precipitation and the pH- $\text{CaCl}_2$  had a highly significant effect on the Al concentration especially when using log-transformed data. Using the original data, the  $\text{NH}_4$  deposition appeared to have the most significant impact on the Al concentration in the topsoil ( $R^2_{\text{adj}} = 54\%$ ) and the subsoil ( $R^2_{\text{adj}} = 46\%$ ). This is illustrated in Fig. 8.21.

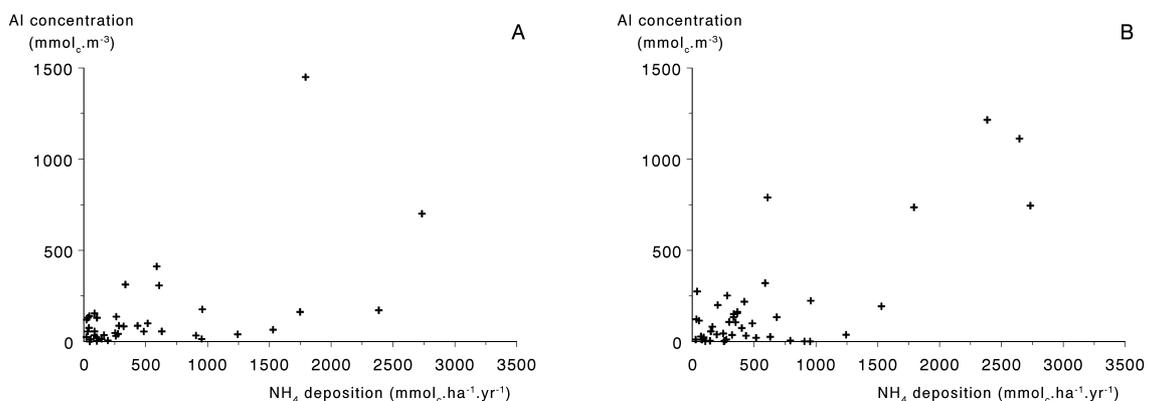


Figure 8.21 Relationship between the  $\text{NH}_4$  deposition and the Al concentration in the topsoil (A) and the subsoil (B) at 42-47 Intensive Monitoring plots.

Using pH-CaCl<sub>2</sub> as a single predictor variable, the explained variation was 28% and 38% only. Using the log-transformed data, however, pH-CaCl<sub>2</sub> appeared to be the most significant predictor for the (annual) average Al concentration, explaining 71% of its variation in the topsoil and 80% in the subsoil. This illustrates the non-linear relationship between Al concentration and pH, that has been discussed before (Section 8.3.1).

The pH was most significantly influenced by the base saturation of the soil, followed by the NH<sub>4</sub> or SO<sub>4</sub>+NO<sub>3</sub> deposition. Using one predictor, base saturation explained approximately 35% of the variation in pH in both topsoil and subsoil, whereas the explanation by either NH<sub>4</sub> or SO<sub>4</sub>+NO<sub>3</sub> deposition varied between 25 and 30%. Combined use of the various predictors led to values of R<sup>2</sup><sub>adj</sub> of 49% in the topsoil and 57% in the subsoil (Table 8.13). Despite the commonly known relationship between pH and base saturation over the whole base saturation range, a large variation in pH values (from less than 4.0 to more than 6.0) appeared to occur at low base saturation (<10-20%) (Fig.8.22). The upper values are quite questionable at this low base saturation.

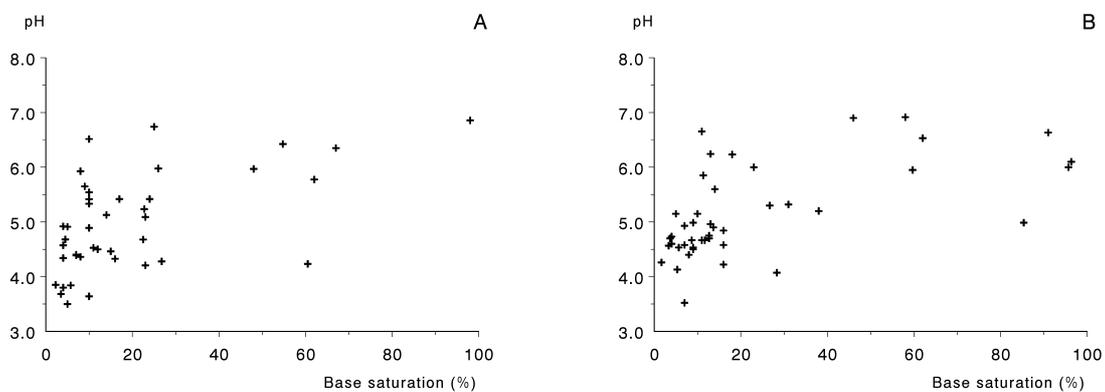


Figure 8.22 Relationship between pH and the base saturation in the topsoil (A) and the subsoil (B) at .. Intensive Monitoring plots.

## 8.4 Conclusions

Soil solution chemistry data at the Intensive Monitoring plots have been evaluated in view of (i) relationships between element concentrations in soil solution, such as Al vs. pH and Al or Ca versus SO<sub>4</sub>+NO<sub>3</sub> in acid soils, (ii) the range in ion concentrations at various soil depths in view of available critical levels in the literature and (iii) the simultaneous impact of atmospheric deposition, meteorological conditions and soil chemistry on the soil solution chemistry. Critical levels were mainly related to NO<sub>3</sub> and Al in view of groundwater pollution and ratios of NH<sub>4</sub> and Al to base cations in view of nutrient imbalances. Major conclusions related to those aspects are given below.

### *Relationships between element concentrations in soil solution*

The Al concentration in both topsoil and subsoil was strongly related to the concentrations of SO<sub>4</sub> and NO<sub>3</sub> at a base saturation below 25% or a pH below 4.5. Above these levels base saturation and pH, the relationship between the concentration of Al and SO<sub>4</sub> plus NO<sub>3</sub> was very weak, indicating that the acidity is neutralised by the release of base cations. The impact of base saturation on the relationship between Ca and strong acid anions was less. However, the

correlation increased and the slope of Ca against  $\text{SO}_4$  plus  $\text{NO}_3$  much more strongly resembled a 1: 1 line at plots with a high base saturation. The logarithmic concentration of Al was clearly related to the pH. Results of the relationship between a calculated logarithmic free (uncomplexed) Al concentration and pH in the organic layer, however, appeared to be much stronger ( $R^2_{\text{adj}} = 74\%$ ) than between the logarithmic total Al concentration and pH ( $R^2_{\text{adj}} = 20\%$ ). The comparison was limited to this layer, because information of DOC, which is crucial to calculate the free Al activity because of the dominating influence of organic Al complexes, was mainly available for the organic layer.

#### *Range in element concentrations in view of critical levels*

Concentrations of  $\text{SO}_4$ ,  $\text{NO}_3$ , total N, Al and Ca was mostly below  $2000 \text{ mmol}_c.\text{m}^{-3}$ , whereas concentrations of  $\text{NH}_4$  were nearly always lower than  $1000 \text{ mmol}_c.\text{m}^{-3}$ . With the exception of Al, the concentrations of those ions decreased significantly going from the organic layer to the mineral topsoil and the mineral subsoil. The increase in Al concentration from the organic layer to the mineral soil, is most likely due to the decrease in water flux in this direction and the dissolution of amorphous Al compounds in the mineral soil. Differences between the mineral topsoil and the mineral subsoil were less significant.  $\text{NH}_4$  most clearly decreased going from the organic layer to the mineral topsoil and the mineral subsoil, indicating the occurrence of preferential  $\text{NH}_4$  uptake and/or nitrification with depth.

The concentrations of  $\text{NO}_3$  and Al exceeded the official ground water quality criterium of  $800 \text{ mmol}_c.\text{m}^{-3}$  at 10-15% of the plots, depending upon the depth considered. The ground water quality criterium of  $20 \text{ mmol}_c.\text{m}^{-3}$  for Al was exceeded at 60% of the plots at greater depth. An Al concentration of  $200 \text{ mmol}_c.\text{m}^{-3}$ , that has sometimes been considered indicative for negative impacts on tree roots, occurs at 25-30% of the plots. As with Al concentrations, the ratios of Al to Ca and the sum of Ca, Mg and K increased with depth. The Al/(Ca+Mg+K) ratios exceeded a critical ratio of 1.0 in approximately 10-15% of the plots, depending on the layer considered. Those percentages are, however, worst case estimates since they are based on measured total Al concentrations instead of free (toxic) Al. Opposite to Al/base cation ratios, the ratios of  $\text{NH}_4$  to base cations decreased with depth. Both the  $\text{NH}_4/\text{K}$  ratio and  $\text{NH}_4/\text{Mg}$  ratio hardly ever exceeded a critical value of 5.0 in the mineral soil.

#### *The simultaneous impact of atmospheric deposition, meteorological conditions and soil chemistry on the soil solution chemistry.*

The variation in concentrations of major ions in the soil solution could to a large extent be explained by differences in atmospheric deposition and to a lesser extent by variations in meteorological conditions (specifically precipitation) and soil chemistry (C/N ratio, pH or base saturation). This followed from the results of multiple linear regression analyses. When using a logarithmic transformation of concentration data, deposition data and meteorological data, the relationships between environmental factors and the concentrations of major ions significantly improved for  $\text{SO}_4$  and  $\text{NO}_3$ , and Al but not for  $\text{NH}_4$ , total N and the base cations Ca, Mg and K. With the exception of  $\text{SO}_4$ , the deposition of  $\text{NH}_4$  had a (highly) significant impact on all the considered compounds, increasing the concentration of N compounds, base cations and Al and decreasing the pH. This can be explained by the acidifying impact of  $\text{NH}_4$  deposition, caused by the conversion of  $\text{NH}_4$  to  $\text{NO}_3$  (nitrification) in the soil. The impact of the deposition of  $\text{SO}_4+\text{NO}_3$  on base cations and Al was generally slightly lower. This result should, however, be interpreted with care, since there is a high correlation between the deposition of  $\text{NH}_4$ ,  $\text{NO}_3$  and  $\text{SO}_4$ .

Using the original data, the explained variation ( $R^2_{\text{adj}}$ ) by environmental factors mostly ranged between 45 and 75%. In all situations, atmospheric deposition of the considered compound was the most important influencing factor with the exception of  $\text{NO}_3$ . For  $\text{NO}_3$  deposition had no significant impact in neither topsoil or subsoil, whereas  $\text{NH}_4$  deposition was highly significant in both cases. In general,  $\text{NH}_4$  deposition was an equally good predictor for the concentration of Ca, Mg and K as the throughfall of the cation itself.

Either the precipitation or the precipitation excess mostly had a significant to highly significant on the concentrations of major ions, especially when using the log-transformed data. In most cases base saturation did not have a significant influence on the Al and base cation concentrations. It did, however strongly affect the pH. Using the log-transformed data, however, pH- $\text{CaCl}_2$  appeared to be the most significant predictor for the (annual) average Al concentration, explaining 71% of its variation in the topsoil and 80% in the subsoil. The influence of the C/N ratio on the measured  $\text{NO}_3$  and  $\text{NH}_4$  concentration was mostly negligible.

## 9 Discussion and conclusions

The major aim of this year's report was to gain more insight in the relationship between atmospheric deposition and soil solution chemistry, which forms the basis for the assessment of input-output budgets, being a prerequisite to derive critical loads for a forest ecosystem. The discussion and conclusions is thus limited to the results of the deposition and soil solution survey. Crown condition of 1996 is only compared with the results of the previous year and results thus obtained are described in Chapter 4 (see also Section 4.4. that summarises the conclusions). Furthermore, results of the forest growth survey have further been interpreted in terms of stand indices that may have an impact on atmospheric inputs. Results thus obtained are described in Chapter 5 (see also Section 5.4. that summarises the conclusions). Information on the first results of the meteorological survey is described in Chapter 7 (see also Section 7.4. that summarises the conclusions).

### *Atmospheric deposition*

Atmospheric deposition data at the Intensive Monitoring plots have been evaluated in view of (i) the relative contribution of wet and dry deposition in the potential acid input, (ii) the relative contribution of N and S compounds and of base cations in the atmospheric input, (iii) the order of magnitude of atmospheric inputs in view of critical loads and (iv) the relationship between atmospheric deposition and stand/site characteristics. Major conclusions related to those aspects are given below.

#### *The relative contribution of wet and dry deposition in the potential acid input*

On average, the contribution of dry deposition appears to be at least half of the total deposition for S and N compounds, whereas it appears to be lower (approximately 30% on average) for base cations. The contribution of wet deposition was dominant in Northern Europe, whereas dry deposition was relatively important in parts of Western and Central Europe.

Comparable results for the contribution of dry deposition were observed by Ivens (1990) using data from nearly 100 plots, concentrated in Northern and Western Europe, where measurements took place between 1967 and 1988. In general, Ivens (1990), however, estimated a larger contribution of dry deposition, especially for the base cations. This may be due to an underestimate of the Na input in throughfall and stemflow at part of the plots. Ratios of this Na input divided by the Na input in bulk deposition below 1.0 do indicate that this does occur. This in turn causes a lower estimated rate of canopy uptake of N, thus underestimating total and dry deposition of N compounds.

#### *The relative contribution of N and S compounds and of base cations in the atmospheric input*

Even though the N to S ratios ranged from approximately 0.3 to 2.0, there was a significant correlation between the input of N and S at the various Intensive Monitoring plots. This correlation will partly be due to the co-occurrence of oil and coal combustion (the major source of SO<sub>4</sub>), traffic (the major source of NO<sub>3</sub>) and animal husbandry (the major source of NH<sub>4</sub>). Another part of the explanation may be the co-deposition of ammonia and sulphur dioxide on water films of the tree surfaces (Adema et al., 1986). The relatively high correlation between NH<sub>4</sub> and SO<sub>4</sub> deposition is an indication for this process.

On average, the total nitrogen input is 50% larger than the total sulphur input. Even though this result may be influenced by the calculated N uptake rates, the comparatively high N deposition at a European wide scale is a striking result. In the eighties, S emissions were generally considered the most important cause of acid deposition. However, since then S emissions and thereby S deposition have constantly decreased over large parts of Europe whereas N emissions mostly stayed constant or even increased. This causes N to be a dominating factor in the acidic input in large parts of Europe.  $\text{NH}_4$  seems the dominating N compound at most of the plots.

A significant relationship was observed between the deposition of Ca and  $\text{SO}_4$  both in bulk and total deposition. The correlation may partly be due to associated emissions of  $\text{SO}_2$  and Ca from smelters and refineries. Generally, the sources of sulphate and base cations in the atmosphere, however, differ substantially. The increased correlation between Ca and  $\text{SO}_4$  input going from bulk to total deposition suggests that the filtering efficiency of forests influences the deposition of both compounds, despite their differences in particle sizes (Ivens, 1990).

#### *Ranges and geographic variation of atmospheric inputs in view of critical loads*

N deposition is specifically larger than S deposition at most of the Intensive Monitoring plots in Western Europe, whereas the reverse is true for Central Europe. Approximately 45% of the considered plots receive an N input above  $1000 \text{ mol}_c \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ , being a deposition level at which the species diversity of the ground vegetation may be at risk. Critical loads related tree health are higher and deposition levels vary from approximately  $1500\text{-}3500 \text{ mol}_c \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ . A comparison with present loads shows that those impacts most likely occur at several plots. At those deposition levels, tree growth may, however, increase in case of N limitation.

Total deposition of both S and N compounds is clearly correlated and varied mostly between  $100 - 3000 \text{ mol}_c \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ . Very high inputs of acidity ( $> 3000 \text{ mol}_c \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ ) occur at approximately 15% of the plots, located in Western and Central Europe. Negative impacts on the forest ecosystem are likely at those plots but a specific comparison of present and critical acid loads is needed to assess the risk of the acid atmospheric input.

#### *Relationships between atmospheric deposition and environmental factors*

Geographic region appears to have a dominant influence on the limited data set of deposition data in combination with the various environmental factors. Atmospheric deposition of all ions is significantly lower in the Boreal regions compared to Western Europe, whereas  $\text{SO}_4$ ,  $\text{NO}_3$  and Ca deposition is significantly higher in the Central/Eastern part of Europe. There is furthermore a highly significant positive correlation of atmospheric deposition and rainfall, except for  $\text{NH}_4$  and K. The deposition of S and N compounds appears to decrease significantly with an increase in altitude. For base cation deposition, the impact of altitude is, however, insignificant.

#### *Soil solution chemistry*

Soil solution chemistry data at the Intensive Monitoring plots have been evaluated in view of (i) relationships between element concentrations in soil solution, such as Al vs. pH and Al or Ca versus  $\text{SO}_4 + \text{NO}_3$  in acid soils, (ii) the range in ion concentrations at various soil depths in view of available critical levels in the literature and (iii) the simultaneous impact of atmospheric deposition, meteorological conditions and soil chemistry on the soil solution chemistry. The evaluation focused on the chemistry of major ions in soil solution impacted by N and S deposition, either directly ( $\text{SO}_4$ ,  $\text{NO}_3$ ,  $\text{NH}_4$ ) or indirectly through soil buffering reactions (H, Al, Ca, Mg, K).

### *Relationships between element concentrations in soil solution*

The Al concentration in both topsoil and subsoil was strongly related to the concentration of  $\text{SO}_4$  and  $\text{NO}_3$  at a base saturation below 25% or a pH below 4.5 indicating that the acidity is neutralised by the release of Al in those acid soils. This result is in line with several other findings in the literature (e.g. de Vries et. al., 1995). The concentration of free (uncomplexed) Al is toxic to roots above a certain critical level. Results showed a very good relationship between the calculated logarithmic free (uncomplexed) Al concentration and the pH in the organic layer. In the mineral layers, this relationship could not be assessed because information of DOC, which is crucial to calculate the free Al activity because of the dominating influence of organic Al complexes was generally not available for those layers. Considering the importance of information on the concentration of free (uncomplexed) Al in view of their potential toxic effects on roots, it seems crucial to measure DOC in the soil solution.

### *Range in element concentrations in view of critical levels*

Concentrations of  $\text{SO}_4$ ,  $\text{NO}_3$ , total N, Al and Ca were nearly always lower than  $2000 \text{ mmol}_c.\text{m}^{-3}$ , whereas concentrations of  $\text{NH}_4$  were nearly always lower than  $1000 \text{ mmol}_c.\text{m}^{-3}$ . The concentration of  $\text{NO}_3$  exceeded the official ground water quality criterium of  $800 \text{ mmol}_c.\text{m}^{-3}$  at 18-26% of the plots, depending upon the depth considered. The Al/(Ca+Mg+K) ratios exceeded a critical ratio of 1.0 in approximately 10-20% of the plots, depending on the layer considered. Results are, however, based on total concentrations of Al, whereas it is specifically the concentration of free (uncomplexed) Al that is toxic to roots. As stated above, an assessment of the free Al was hardly possible for the mineral layer, due to missing information on DOC. It is thus advised that DOC is always measured to allow such calculations. Both the  $\text{NH}_4/\text{K}$  ratio and  $\text{NH}_4/\text{Mg}$  ratio hardly ever exceeded a critical value of 5.0 in the mineral soil.

### *The simultaneous impact of atmospheric deposition, meteorological conditions and soil chemistry on the soil solution chemistry.*

The variation in concentrations of major ions in the soil solution could to a large extent be explained by differences in atmospheric deposition and to a lesser extent by variations in meteorological conditions (specifically precipitation) and soil chemistry (C/N ratio, pH or base saturation). In most cases base saturation did not have a significant influence on the Al and base cation concentrations. It did, however strongly affect the pH. Using the log-transformed data, however, pH- $\text{CaCl}_2$  appeared to be the most significant predictor for the (annual) average Al concentration.

Unlike results presented by Dise et al. (1998) and Gundersen et al. (1998), the influence of the C/N ratio on the measured  $\text{NO}_3$  and  $\text{NH}_4$  concentration was mostly negligible. In case of  $\text{NH}_4$  in the subsoil there was a significant effect, but this is likely to be an artefact since the 'sign' is opposite to what is expected (a '+' sign, which implies that the  $\text{NH}_4$  concentration increases with an increase in the C/N ratio). The small to insignificant influence of the C/N ratio on  $\text{NO}_3$  and  $\text{NH}_4$  concentrations was also observed in 150 forested stands in the Netherlands (De Vries and Leeters, 1999). A significant relationship was only found in a subset of well-drained soils with coniferous stands. A more in-depth analysis, based on much more data is necessary to further investigate the possible role of the C/N ratio of the soil on the N dynamics

### ***Overall conclusions and final discussion***

- On average, the total nitrogen input is 50% larger than the total sulphur input. N deposition is larger than S deposition at most of the Intensive Monitoring plots in Western Europe, whereas

the reverse is true for Central Europe. Total N inputs exceed a deposition level of 1000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> at approximately 45% of the plots. At those plots, adverse impacts on species diversity of ground vegetation are likely.

- Total deposition of acidity, caused by both S and N compounds varied mostly between 100 – 3000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. Very high inputs of acidity (> 3000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>) occur at approximately 15% of the plots, located in Western and Central Europe. Negative impacts on the forest ecosystem are likely at those plots
- Concentrations of SO<sub>4</sub> and NO<sub>3</sub>, which are mainly influenced by S and N deposition, are significantly related to Al concentrations in acid soils. NO<sub>3</sub> concentrations in the subsoil exceed official groundwater quality criteria at 18% of the plots. Molar ratios of Al to base cations above critical levels (levels indicative for adverse effects on roots) occur at some 10 – 20% of the plots.
- Apart from the geographic region, atmospheric deposition is significantly influenced by altitude, tree height and rainfall. The variation in concentrations of major ions in the soil solution could to a large extent be explained by differences in atmospheric deposition and to a lesser extent by variations in meteorological conditions (specifically precipitation) and soil chemistry (especially pH or base saturation in relation to Al and to a lesser extent base cations).

It has to be noted that the conclusions are based on a limited number of plots both for atmospheric deposition and soil solution chemistry. This is especially true with respect to the studies relating soil solution chemistry, and to a lesser extent atmospheric deposition, to environmental factors. This certainly biases the results. In most cases, the results are, however, in line with knowledge available from the literature.

Care should also be taken with the information about the percentages of plots exceeding certain critical values. Regarding the use of groundwater quality criteria, one has to be aware that NO<sub>3</sub> concentrations are generally lower in groundwater than in soil solution draining to ground water, due to the occurrence of denitrification. Furthermore, the critical Al/BC ratios are mostly based on laboratory studies with seedlings and not on actual field data (Sverdrup and Warfinge, 1993). The percentages are more an indication of potential problems.

The results of the various regression relations do indicate the possibility of upscaling response variables such as soil solution chemistry to e.g. level 1 plots. This is, however, only relevant or acceptable when (i) the relationships do explain a large part (e.g. more than 60%) of the variation in the response variable and (ii) the predictor variables are all available at the level 1 plots or can be estimated with a reasonable accuracy. The reasonable to good comparison of site specific and interpolated precipitation data at various Intensive Monitoring plots indicates that this variable can be used for predictive purposes. A comparison of measured and modelled atmospheric deposition data is further needed to investigate the possibility of upscaling the results.

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## **Annex 1 A comparison of results obtained by zero tension lysimeters, suction cups and centrifugation in different studies.**

In theory, the chemical composition of the different fractions of the soil solution reflects different processes taking place in the soil, thus leading to problems when attempting to make direct comparisons between soil solution data obtained by different techniques. Zero-tension and suction cup lysimetry are relatively non-destructive methods, and changes in soil solution quality can usually be followed at the same point for decades. Soil centrifugation, on the other hand, is a destructive technique since soil samples have to be taken. All methods require a considerable number of replications in order to obtain representative results. In contrast to the lysimeter techniques, the samples for soil centrifugation cannot be taken from exactly the same points in successive samplings.

Below, we further concentrate on differences between suction cup and zero-tension lysimeters and on differences between the destructive centrifugation method and the non-destructive lysimeter methods.

### ***Comparison of suction cup and zero-tension lysimeters***

The choice of lysimeters to collect soil solution is generally dependent on the properties of the soil under investigation. The use of zero-tension lysimeters is ideal in soils composed of sorted sand or gravel (e.g. Lumme et al., 1995). Zero-tension lysimeters are often used if the aim of the study is to determine, in addition to percolation water quality, water and element fluxes on an areal basis (e.g. Helmisaari, 1995). Sometimes it is, however, impossible to install zero-tension lysimeters in very stony soils due to their relatively large size compared to suction cup lysimeters. In theory, fluxes can also be calculated using suction cup lysimeters on the basis of element concentrations in the soil water and the water flux provided by water flux models. One advantage of the suction cup lysimeters is that soil water can often be sampled during dry summer months when there is no vertical movement of water in forest soils. In interpreting the results of early monitoring data, it is further important to realise that installation of lysimeters may cause disturbances in the cycling of elements in the forest soil, for example due to increased leaching of dissolved organic matter. This is a significant problem in the installation of several types of zero-tension lysimeter. During lysimeter installation, the plant roots are cut, resulting in the acceleration of mineralisation processes in the forest soil and subsequent changes in percolation water quality (Lundström, 1993).

Zero-tension lysimeters only collect water that percolates down through the soil profile as a result of gravitational forces (percolation water), i.e. when the moisture content of the overlying soil layers exceeds field capacity. Suction-cup lysimeters sample that fraction of the soil solution which is held in the spaces between soil particles by capillary forces, as well as percolation water if the soil moisture content exceeds field capacity. The actual fraction which is sampled depends on the strength of the vacuum applied, and on the hydraulic tension of the soil in question.

Interactions between soil solution and the material used to construct the lysimeters may furthermore influence the results. This is especially the case with suction cup lysimeters, where the adsorption or desorption of certain elements in or on the lysimeter may affect soil water quality (Rasmussen et al., 1986). These processes are dependent on the suction-cup materials used: porous cups are usually made of teflon, porcelain or sintered glass. In addition, the pore size of the suction-cups may have an influence on soil water quality. For example, fine-porous (<

1µm) ceramic samplers may interact with phosphate, heavy metals and humic substances in the soil solution and give erroneous results (Environment Data Centre, 1993).

In the following, we report some results of a comparison of suction cup and zero tension lysimeters used to obtain soil solution at five Intensive Monitoring plots in Finland (Derome and Lindroos, 1997). Results of a comparison between zero tension and suction cup lysimeters at 20 cm depth for four plots (Table A1) show remarkable differences for nearly all ions. These results, which refer to concentrations of cations and anions in water samples obtained during the 1996 growing season, are only meant to illustrate the possible differences.

*Table A1 Mean pH, total Al, Ca, Mg, K, Na, NH<sub>4</sub>-N and SO<sub>4</sub>-S concentrations in soil solution obtained using zero tension lysimeters and suction cup lysimeters at a depth of 40 cm at four Intensive Monitoring plots in Finland in 1996. The standard error of the mean is given below each value.*

Plot nr	lysimeter	N <sup>1)</sup>	pH	Concentration (mg.l <sup>-1</sup> )						
				Al	Ca	Mg	K	Na	NH <sub>4</sub> -N	SO <sub>4</sub> -S
10	zero	6	4.74	0.52	0.96	0.39	1.20	0.63	0.07	2.09
	tension		(0.27)	(0.18)	(0.42)	(0.16)	(0.57)	(0.28)	(0.05)	(1.36)
	suction	8	6.26	0.04	0.96	0.45	2.06	1.68	0.96	2.00
	cup		(0.36)	(0.04)	(0.32)	(0.19)	(3.89)	(0.34)	(0.32)	(0.32)
11	zero	6	5.16	0.28	2.47	0.35	0.09	1.68	0.02	3.77
	tension		(0.41)	(0.25)	(1.22)	(0.14)	(0.02)	(0.26)	(0.03)	(1.37)
	suction	14	5.56	0.31	2.76	0.58	0.07	2.21	0.00	4.33
	cup		(0.35)	(0.14)	(2.18)	(0.24)	(0.03)	(0.50)	(0.01)	(2.13)
12	zero	8	5.08	0.54	1.26	0.72	0.25	2.79	0.09	3.65
	tension		(0.56)	(0.42)	(0.46)	(0.39)	(0.22)	(0.38)	(0.11)	(0.66)
	suction	28	5.33	0.48	1.96	0.21	0.21	3.64	0.15	9.65
	cup		(0.58)	(0.42)	(0.29)	(0.12)	(0.12)	(0.67)	(0.09)	(4.78)
13	zero	6	5.08	0.28	2.50	0.39	0.46	0.88	0.18	4.52
	tension		(0.38)	(0.13)	(1.57)	(0.12)	(0.15)	(0.50)	(0.08)	(2.10)
	suction	6	6.17	0.62	3.13	0.61	0.44	1.44	0.15	6.71
	cup		(0.47)	(0.06)	(0.33)	(0.08)	(0.18)	(0.29)	(0.11)	(1.37)

<sup>1)</sup> N = number of observations

Most striking results are the systematically lower pH, Na and SO<sub>4</sub> concentration in percolation water obtained by zero-tension lysimeters, compared to soil water obtained by suction cups. Similar results were obtained at 20 cm depth (Derome and Lindroos, 1997). Concentrations of Ca, Mg and K were also mostly lower in zero-tension lysimeters, but sometimes higher concentrations were measured at 40 cm depth. Comparable results were found at 20 cm depth, except for K concentrations which were consistently much higher in zero-tension lysimeters. Except for one plot (plot 10), NH<sub>4</sub> concentrations were quite comparable in the zero-tension lysimeters and suction cup lysimeters (Table A1). At 20 cm depth, however, NH<sub>4</sub> concentrations tended to be higher in zero-tension lysimeter.

Derome and Lindroos (1997) interpreted the differences partly in terms of different interactions of the percolation/soil water obtained and the soil matrix. The consistently lower pH values in zero-tension lysimeters do indeed suggest less interaction of percolation water with the soil matrix. The same is true for the much higher K concentrations at 20 cm depth, which may result from K cycling. With respect to Na, the significantly higher concentration in the suction cups is presumably due to interaction with the collector bottles, that were made of sodium borosilicate glass (Derome and Lindroos, 1997).

In summary, it can be stated that differ the calculation of element budgets is certainly influenced by the type of lysimeter used, specifically at lower soil depths.

### ***Comparison of suction cup lysimeters and centrifugation***

Unlike the use of lysimeters, centrifugation is a destructive method that is not specifically suited for regular (e.g. weekly or monthly) monitoring. Instead it is used in the Netherlands at an annual interval. The main advantage above lysimeters is that an average value for a plot at a given period can more easily be obtained by using a pooled sample.

There are also differences to be expected between lysimeters and centrifugation. In general, the equivalent suction that can be applied by centrifugation ( $\approx 1000$  kPa) is much higher than by vacuum extraction using suction cups ( $< 100$  kPa). The suction (gravity forces) applied depends on the centrifugation speed and the angle of the rotor. This implies that the centrifugation of fresh soil samples results in the collection of soil solution that is more strongly bound to the soil matrix than suction cups. In coarse sandy soils, this may not cause much difference, but in more loamy soils there is a clear difference in the concentration in pores within soil particles than between soil particles. The latter pores mainly contain percolation water filtering down the soil profile (see before). Zabowski and Ugolini (1996) thus conclude that centrifugation can best be used to determine the nutrient availability, whereas lysimeters (preferably zero tension) can best be used to calculate element fluxes through the profile.

Insight in the difference in ion concentrations obtained by centrifugation and suction cup lysimeters can be derived from a comparative study of both methods at two forested plots (Speuld and Ysselstein) in the Netherlands (Verhagen and Diederien, 1991). In this study, suction cup lysimeters were installed at fifteen spots that were situated two meters apart at two depths (10 or 20 cm and 40 or 60 cm). Soil samples for centrifugation were taken as close as possible to the lysimeters to minimise impacts of spatial variability. Results of the study (Table A2) showed that ion concentrations are generally significantly higher in the centrifugates than in the suction cup lysimeters with the exception of pH, Al and  $\text{NH}_4$ . On average, Al concentrations were even lower in centrifugates. The Al/Ca ratio, which is an important indicator for stress induced by acidification (e.g. Ulrich and Matzinger, 1983), was therefore significantly higher in suction cups than in centrifugates. Comparable results were found by Zabowski and Ugolini (1990).

*Table A2 Comparison of mean and median element concentrations ( $\text{mmol}_e\text{m}^{-3}$ ) in lysimeters and centrifugates of 60 soil samples at two forested plots in the Netherlands*

Element	Mean concentration		Median concentration		Significance
	lysimeters	centrifugates	lysimeters	centrifugates	
pH	3.5	3.5	3.3	3.4	-
Al	697	658	485	511	-
Ca	308	540	291	484	+
Mg	132	174	123	148	+
K	89	123	49	109	+
Na	570	655	522	544	+
$\text{NH}_4$	472	503	158	190	-
$\text{NO}_3$	689	776	689	793	+
$\text{SO}_4$	935	1100	881	1000	+
Cl	985	1074	854	945	+
Al/Ca	2.4	1.0	1.1	0.7	+
$\text{NH}_4/\text{K}$	3.9	3.4	2.0	2.1	-

+ = significant difference according to Wilcoxin test

In the sandy soils studied, the differences are not very large. Results are all in the same order of magnitude. Except for Ca, differences between mean and median values obtained by suction cups and centrifugates vary mostly between 10 and 20%. As stated before, the differences may, however, be larger in loamy and clayey soils.

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