## 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** 

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

Technical Report 2002

Prepared by: Forest Intensive Monitoring Coordinating Institute, 2002



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**Technical Report 2002** 

W. de Vries G.J. Reinds H. van Dobben D. de Zwart D. Aamlid P. Neville M. Posch J. Auée J.C.H. Voogd E.M. Vel

Forest Intensive Monitoring Coordinating Institute, 2002

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# Abstract

EC - UN/ECE, 2002; W. de Vries, G.J. Reinds, H. van Dobben, D. de Zwart, D. Aamlid, P. Neville, M. Posch, J. Auée, J.C.H. Voogd, E.M. Vel. *Intensive Monitoring of Forest Ecosystems in Europe, 2002 Technical Report.* EC, UN/ECE 2002, Brussels, Geneva, 175 pp.

Apart from an overview of the implementation of the Pan-European Intensive Monitoring Programme of Forest Ecosystems up to 1999, this year's report focuses on plant biodiversity and critical loads for nitrogen, acidity and heavy metals. Major conclusions with respect to relationships between species diversity of the ground vegetation and environmental factors are:

- For a limited number of species, there is a significant relationship between the occurrence probability and soil pH, with most species favouring alkaline conditions and few species being more prominent under acid conditions. A relationship between occurrence probability and atmospheric nitrogen deposition was found for very few individual species only.
- Approximately 40% of the variation in species numbers can be explained by environmental factors. The pH in the organic layer explains most of the variation, followed by tree species, soil factors, climate and atmospheric deposition.
- Approximately 20% of the variation in the abundance of the various species in the ground vegetation could be explained by the actual soil acidity, tree species and climate in terms of precipitation and temperature. The variation in bulk deposition chemistry also explained a small part of the variation. The weak effect of the 'deposition' predictors is only based on the spatial pattern of both vegetation and predictors and may partly be hidden in the effect of actual soil acidity. A stronger effect of deposition on vegetation development in time is expected.

Major conclusions with respect to critical loads for nitrogen, acidity and heavy metals and their exceedances by present loads are:

- At approximately 50% of the investigated plots, critical nitrogen loads related to impacts on ground vegetation and on the vitality of coniferous trees are exceeded by the present deposition. At these plots the risk for drought stress, frost, pests and diseases is increased and additionally the species diversity of the ground vegetation might be endangered. At approximately 90% of the plots, it is likely that N is accumulating in the ecosystem. This conclusion holds for the evaluated 234 plots, which mainly occur in Central Europe with an average nitrogen deposition of 19 kg.ha<sup>-1</sup>.yr<sup>-1</sup>.
- At 33% to 64% of the investigated plots, the critical acid loads are exceeded, depending on the critical limits used. At these plots the functioning of tree roots is endangered and additionally base cations or aluminium might be leached from the soil. This conclusion holds for the evaluated 226 plots with an average acid (nitrogen plus sulphate) deposition of approximately 2100 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>.
- Highest exceedances of critical loads, do occur in central Europe and for nitrogen also in western Europe where present loads are high and critical loads are relatively low.
- Critical loads of cadmium and lead were exceeded on 78-91% of the plots for lead and on 6-29% of the plots for cadmium, depending on the critical limit used for the dissolved metal concentration and using modelled deposition data. At these loads, some impact may occur on the soil fauna and plants, but the results are based on very stringent criteria and do refer to the occurrence of a steady-state situation.

Keywords: Intensive monitoring, forest, ground vegetation, biodiversity, atmospheric deposition, critical loads, acidification, eutrophication, heavy metals.

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# Preface

The 'Pan-European Programme for Intensive and Continuous Monitoring of Forest Ecosystems' has been implemented to gain a better understanding of the effects of air pollution and other stress factors on forests. At present 866 permanent observation plots for Intensive Monitoring of forest ecosystems have been selected (510 in the European Union and 356 in several non-EU countries). The Intensive Monitoring Programme includes the assessment of crown condition, increment and the chemical composition of foliage and soil on all plots, whereas atmospheric deposition, meteorological parameters, soil solution chemistry and ground vegetation composition are monitored at selected plots. Data are submitted to the Forest Intensive Monitoring Co-ordinating Institute (FIMCI), being a contractor of the European Commission (EC). FIMCI, which is a joint initiative of Alterra Green World Research and Oranjewoud International, has been set up to validate, store, distribute and evaluate the data at European level. Apart from the data management, FIMCI also acts as an information centre for National Focal Centres (NFC's), of both EU-Member States and the other participating countries of ICP-Forests.

Between 1997 and 2000, four reports have been published with results from (nearly) all the surveys carried out: crown condition, soil chemistry, foliar chemistry, forest growth, atmospheric deposition, meteorology and soil solution chemistry. Results focused on relationships between crown condition, soil and soil solution chemistry and foliar chemistry on one hand and atmospheric deposition and meteorology on the other hand, using statistical techniques for interpretation. Since 2001, certain topics are highlighted by more in-depth studies. The focus of last year's report was on water and element fluxes through the forest ecosystem. The report of 2001 also contained first data on the species diversity of the ground vegetation, focusing on data assessment methods and data comparability and a presentation of the results.

The focus of this year's report is on: (i) relationships between both the occurrence probability of individual species and the species composition of ground vegetation and environmental factors and (ii) critical loads for nitrogen, acidity and heavy metals for the forest ecosystem in comparison to element inputs from the atmosphere. This focus is a logic follow up of last years report. As with that report, external experts have been involved in writing this report.

The target groups of this report are the active participants of the Intensive Monitoring Programme (National Focal Centres, National Involved Research Institutes, Scientific Advisory Group, 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 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 stress factors on forest ecosystems. At present, the programme covers 866 selected plots in 30 participating countries (510 plots in the EU and 366 plots in non-EU countries). In total 791 Intensive Monitoring plots have been installed. Some surveys are mandatory and have to be carried out on all plots (crown condition, soil chemistry, foliage and forest growth). At part of the plots, assessments of atmospheric deposition (499 plots), meteorology (202 plots), soil solution chemistry (243 plots), ground vegetation (632 plots) and remote sensing (approximately 155 plots) are carried out. For most of the plots (around 85%) information on the methods applied is available. The results up to 1999 include data for 765 plots with respect to crown condition, 751 plots for foliar composition, 694 plots for soil, 583 plots for forest growth, 515 plots for atmospheric deposition, 674 plots for ground vegetation, 242 plots for soil solution and 177 plots for meteorology. Furthermore, data on ambient air quality and phenology are available at a limited number of plots.

## **Objectives**

The aim of the thematic Technical Report is to inform policymakers and scientist with relevant information from the monitoring programme and to promote co-operation between FIMCI and other (potential) users of the data. It includes the scientific background of the major results presented in the executive report, that is specifically aimed at both policy makers and the wider public. This years report is the second in the series and focuses on:

- Relation between plants species composition and environmental factors
- Critical loads for nitrogen, acidity and heavy metals and their exceedances by present loads

## Species composition of the ground vegetation

#### Approach

After the UNCED conference in Rio de Janeiro in 1992, there is a growing concern over the world-wide loss of biodiversity. The species composition of the ground vegetation, which is assessed at Intensive Monitoring plots, is an indication of the floristic biodiversity of forest ecosystems thus contributing to information on biodiversity in European forests. Presently, ground vegetation data are available for more than 650 plots. Combined with the data from the same plots on soil conditions and deposition the vegetation survey offers a unique opportunity to analyse and understand the relation between the species composition of the ground vegetation and environmental factors. However, temporal changes in plant communities can only be analyse after a repetition of the surveys with a five year interval.

Preliminary evaluations demonstrate that:

- Species numbers show a slight North-South gradient with increasing species numbers in the Mediterranean areas.
- The species diversity index according to Simpson shows large, rather random differences in species diversity between plots within a country, from which only a very slight North-South gradient can be detected.

- Ellenberg indicators for temperature and soil acidity confirm the geographical differences in temperature and acidity. With repeated surveys, trends in these Ellenberg indicators will be an important issue.

## Relationships between the occurrence probability of species and environmental factors

Derived relationships between the occurrence probability of individual species and environmental factors for 332 different plant species allow the following conclusions:

- The median explained deviance of the models constructed for all individual species is about 30%. The deviance varies mostly between 10 and 70%. This means that the variation in the occurrence probability can sometimes be explained very well by the included environmental factors (up to 70%), but sometimes rather poor (as low as 10%).
- For a limited number of species (36), there is a significant relationship between the occurrence probability and soil pH, with most species favouring alkaline conditions and few species being more prominent under acid conditions.
- A relationship between occurrence probability and atmospheric nitrogen deposition was only found for a very few individual species, some favouring nitrogen rich and some nitrogen poor circumstances.

## Relationships between the species composition of ground vegetation and environmental factors

Derived relationships between the species composition of ground vegetation and environmental factors, related to soil, tree species, climate and atmospheric deposition, allow the following conclusions:

- Approximately 40% of the variation in species numbers can be explained by environmental factors, whereas the explanation of the Simpson index is approximately 15%. The pH in the organic layer explains most of the variation, followed by tree species, soil factors related to nutrient availability, climate and atmospheric deposition.
- Approximately 20% of the variation in the abundance of the various species occurring in the ground vegetation could be explained by the included environmental factors. As with the species numbers and the Simpson index, the explained variance is almost exclusively due to the actual soil situation, tree species and climate, which contribute in approximately equal amounts to the fit of the model. Only a small portion of the explained variance is due to deposition chemistry. Depending on the type of analysis carried out, Na, K, NH<sub>4</sub> and NO<sub>3</sub> are the ions in deposition that are significant, and of these two only NH<sub>4</sub> and NO<sub>3</sub> are of anthropogenic origin.
- The explanation increases with 13% when country is included as an explicit predictor, but this only illustrates that part of the variation can be explained by differences in data assessment methods.

Although the present results indicate a strong effect of the 'traditional' predictors for ground vegetation and a weak effect of the 'deposition' predictors, it should be stressed that this conclusion is only based on the spatial pattern of both vegetation and predictors. In interpreting the low percentage variance explained by the deposition terms, it should be kept in mind that the total variance in the present dataset is extremely large, as it covers forests of all climate zones and soil types over Europe. Therefore the effect of climate and soil is far larger than the effect of deposition. Rather, the effect of deposition should be considered as a weak 'signal' that is to be separated from large amount of 'background noise' caused by the traditional factors. In this view, it is already a clear signal that a significant effect of deposition is found anyway. Only in repeated measurements the 'background noise' is cancelled out, and the effect of (a change in) deposition can be determined with more certainty.

It may thus still be possible that there is a strong effect of deposition on vegetation in the temporal domain, for example that nitrogen-demanding species show a strong increase in places where deposition is high. However, the determination of such relations is outside the scope of the present study, and will only become possible when sufficient repetitive measurements are available. By continuation of this survey, ICP Forests will have data not only on distribution but also on any change in plant community over the past 5 years. This will allow a study on impacts of environmental factors on temporal changes, probably within 2 - 4 years.

## Critical loads for nitrogen and acidity and their exceedances

This report presents a European wide assessment of critical loads for Intensive Monitoring plots in comparison to present loads. The critical loads were calculated for approximately 230 plots, where all relevant available data on deposition, meteorology, forest growth and soil and soil solution chemistry were available. So far most countries in Europe have made critical load maps for nitrogen and acidity for forests based on estimated data on tree uptake, soil weathering and nitrogen retention.

## Approach

A comparison of present element inputs from the atmosphere and critical loads give insight into the sites that are potentially at risk. Critical loads refer to the deposition loads of air pollutants (SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub> and metals), below which no adverse effects on ecosystems are expected in a steady-state situation. Critical loads for nitrogen and acidity were assessed with steady-state soil models, by calculating the deposition loads which avoid the violation of a chemical criterion in a steady-state situation. Different types of critical loads were calculated, related to impacts on:

- Soil: In this context, no further net accumulation of nitrogen or loss of exchangeable base cations or readily available aluminium in the forests soil was accepted (stand-still principle).
- Species diversity of the ground vegetation: Here it was required that concentrations of dissolved nitrogen stay below critical limits in soil solution (effect that was only considered in view of the eutrophying impact of nitrogen).
- Trees: This includes risks for drought stress, frost, pest and diseases induced by nitrogen input for conifers and negative impacts on root uptake as a result of increased acidity. For the first effect, critical limits were used for the N content in needles of conifers and for the second effect, critical ratios of toxic aluminium to base cations were applied.

## Critical nitrogen loads and their exceedances

The average present nitrogen load on the investigated 234 plots was nearly 20 kg.ha<sup>-1</sup>.yr<sup>-1</sup>. Lowest loads were found for pine, followed by spruce, reflecting their location in mostly low deposition areas, such as Scandinavia.

Critical N loads for soil, which aim at no further net accumulation of nitrogen, were calculated by requiring a natural very low N leaching rate from the system. This leads to low critical nitrogen N loads, which ranged mostly between 2 and 14 kg.ha<sup>-1</sup>.yr<sup>-1</sup> with an average near 8 kg.ha<sup>-1</sup>.yr<sup>-1</sup>. Values were clearly lower for pine, with a lower N uptake, than for the other tree species. This critical N load was exceeded at 92% of the plots. This does not imply, however, that at 92% of the plots are at risk because of impacts on tree condition or ground vegetation. Instead, it implies that at most of the plots, N is accumulating in the soil system. It means that the soil N pool is increasing (in some situations only with an extremely low speed) and this may ultimately (years, decades or centuries) lead to unacceptably high concentrations.

Critical N loads related to impacts on the ground vegetation were calculated by assuming an acceptable limit for N in soil solution of 0.2  $\text{mol}_{c.m^{-3}}$  (2.8 mgN.l<sup>-1</sup>). This lead to an average critical N load of 17 kg.ha<sup>-1</sup>.yr<sup>-1</sup> and a median value of 13 kg.ha<sup>-1</sup>.yr<sup>-1</sup>. These results are in line with empirical data, which vary mostly between 7 and 20 kg.ha<sup>-1</sup>.yr<sup>-1</sup> with an average near 14 kg.ha<sup>-1</sup>.yr<sup>-1</sup>. These loads were exceeded at 58% of the plots. This comparison shows that changes in plant biodiversity are likely in large parts of the European forests.

Critical N loads related to impacts on trees were limited to conifers, which aimed at concentrations of nitrogen in the foliage below a critical limit were only calculated for conifers. A content of 18 g.kg<sup>-1</sup> was used as critical level; above this limit the risk for drought stress, frost, pest and diseases increases. For deciduous trees like oak and beech the relation between N deposition and foliar N concentration is not so clear and no critical limits are defined. The average critical N load thus calculated was near 14 kg.ha<sup>-1</sup>.yr<sup>-1</sup> for pine and near 20 kg.ha<sup>-1</sup>.yr<sup>-1</sup> for spruce. This load was exceeded at 45% of the plots with conifers.

#### Critical acid loads and their exceedances

The average present acid load on all investigated 226 plots was nearly 2100 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. As with nitrogen, lowest loads were found for pine, followed by spruce. Critical loads for soil were calculated by requiring no further loss of exchangeable base cations in base rich forests soil (loess, clay and peat soils) and no further loss of readily available aluminium in base poor sandy forest soils. The effect–based critical loads were calculated by aiming that ratios of toxic aluminium to base cations in the soil solution stayed below a critical limit of 0.8 for pine and spruce and 1.6 for oak and beech.

The critical acid loads for soil, which were on average approximately 1600 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>, were exceeded at 64% of the plots. The critical acid loads for trees (impacts on root uptake) were approximately twice as high. Values ranged mostly between 500 and 8000, with an average near 3500 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. Critical acid loads were clearly lower for pine and spruce, which are more sensitive to aluminium, than for oak and beech. Considering all plots, critical loads were exceeded at 33% of the plots when impacts on tree roots are considered This is in line with measurements of aluminium to base cations ratios in the soil solution and shows that impacts on forests are likely in European forests.

#### Geographic variation in critical loads and their exceedances

The geographic variation in the exceedance of critical loads<sup>1</sup> is large for both nitrogen and acidity. Highest exceedances do occur in Western and Central Europe where present loads of both nitrogen and sulphur are generally high and critical loads are relatively low. The geographic variation in critical loads with respect to eutrophication impacts on the ground vegetation (nitrogen) and acidification impacts on the trees (acidity) shows that high critical nitrogen loads (>1000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>) mainly occur in Central and Southern Europe, mainly due to high N uptake, specifically by broadleaves. Critical nitrogen loads were calculated to be much smaller in northern Europe, where the net uptake of N by trees is low, but also in parts of Southern Europe due to low N uptake rates (impact of drought stress) and low precipitation excesses . In general, the critical acid load also increased from the northern boreal regions to Southern Europe, due to

<sup>&</sup>lt;sup>1</sup> the difference between present deposition load and critical load

an increase in base cation input from the atmosphere and by soil weathering and in nitrogen uptake.

#### Present deposition thresholds of nitrogen and acidity and their exceedances

Since calculation of a critical load assumes a steady-state situation, an excess of those loads does not necessarily imply that the forest ecosystem is at risk yet. Apart from critical loads, present deposition thresholds were therefore calculated, being the deposition levels that lead to risks to the forest at present and not in a steady-state situation. It are the loads causing concentrations of nitrogen and acidity in soil solution that presently violate critical limits. Present deposition thresholds (PDT) for nitrogen and acidity did often strongly deviate from the long-term critical loads (CL). In plots where the present concentrations exceeded the critical limits, PDT was lower than CL and often became negative (it is impossible to attain a critical limit within one year), whereas in the opposite situation, the PDT was generally much larger than the CL.

The present deposition thresholds related to tree impacts, in terms of elevated N concentrations in the foliage of conifers were generally much larger than the steady-state critical loads and consequently the exceedance was much lower. The calculated exceedance for the 72 considered plots was only 15%, compared to 45% of the 164 considered plots in the critical load calculation. Inversely, the PDT's related to tree root impacts, in terms of elevated dissolved Al/Bc ratios, were generally lower than the CL values and consequently, the area exceeding the critical loads was higher. This implies that the present situation may be worse than the steady state situation, which is most likely due to net release of sulphur in many plots with previous high sulphur inputs. The results confirm the present non steady state situation for nitrogen in terms of N accumulation and for acidity in terms of acidity (sulphur) release.

#### Critical metal loads and their exceedances

Concern about the atmospheric input of heavy metals (specifically cadmium and lead) to forest ecosystems is specifically related to the impact on soil organisms and the occurrence of bioaccumulation in the organic layer. Copper and zinc are essential elements, and deficiencies are relevant for forest growth and forest health. Heavy metals lead (Pb), cadmium (Cd), copper (Cu) and zinc (Zn) are measured at part of the plots.

#### Approach

The critical loads were calculated by aiming at (i) no further net metal accumulation in the forest soil (stand-still principle) or (ii) that metal concentrations in soil solution in a steady-state situation stay below critical limits related to effects on plants and soil organisms (effect-based principle). Results were limited to lead (Pb), cadmium (Cd), copper (Cu) and zinc (Zn). Effect-based critical loads were calculated for approximately 240 plots. Two sets of critical limits were used. One based on a large number of indirectly derived data for micro-organisms, soil invertebrates and plants and one on limited measured data for plants. Stand-still loads could be calculated for a limited number of plots with data on metal contents in the mineral topsoil. Furthermore, present Cd, Cu, Pb and Zn deposition could be calculated for a limited number of Level II plots only (for Cd and Pb near 50). In evaluating the results, a comparison was made between:

- Effect-based critical loads and present loads derived from model calculations at all plots
- Stand-still loads and measured deposition at a limited number of plots.

## Critical metal loads and their exceedances

On average the present Pb deposition is much higher than the critical load. When related to the impact on soil fauna (with rather stringent and uncertain limits) exceedances were found on 91% of the plots. Critical load calculated from critical limits for plants were exceeded at 78% of the plots. For Cd, the difference between modelled deposition and critical load is small, with exceedance on 29% of the plots. The modelled Cu deposition only exceeds the critical load in 8% of the plots. Critical loads for plants show an inverse result, with exceedances in only 6% of the plots for Cd, but in 51% of the plots for Cu. This shows the extreme sensitivity of the results tot the value of the (uncertain) critical limit.

High critical metal loads mainly occur in high precipitation areas, such as parts of the UK, Norway and the mountainous areas in Central Europe. It has to be noted that the critical loads are related to a steady-state situation. Critical Cd loads are mainly exceeded in plots in Western and Central Europe, where the present loads metals are relatively high.

# **1** Introduction

In order to gain a better understanding of the effects of air pollution and other stress factors on forest ecosystems, the Pan-European Programme for Intensive and Continuous Monitoring of Forest Ecosystems was established. This chapter first presents information on the background and current status of the Intensive Monitoring Programme (Section 1.1). It then explains 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).

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

## Background of the programme

The Pan-European Programme is based on both the European Scheme on the Protection of Forests against Atmospheric Pollution 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). In 1994, the Intensive Monitoring Programme was established by the EC with the aims to (ICP Forests, 2000):

- Monitor effects of anthropogenic (in particular air pollution) and natural stress factors on the condition and development of forest ecosystems in Europe.
- Contribute to a better understanding of cause-effect relationships in forest ecosystems functioning in various parts of Europe.

At present 862 permanent observation plots for Intensive Monitoring of forest ecosystems have been selected (512 in the European Union and 350 in several non-EU countries). Details on the plots and assessments can be found in chapter 2.

The Intensive Monitoring Programme includes the assessment of crown condition, forest growth (increment) and the chemical composition of foliage and soil on all plots. Additional measurements on selected plots include atmospheric deposition, meteorological parameters, soil solution chemistry and ground vegetation. Within each of these surveys, a number of mandatory and optional parameters has been defined. The temporal resolution of the present surveys is scheduled as follows:

- Crown condition (at least once a year)
- Chemical composition of the concentrations 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 and phenology (continuous)
- Ground vegetation (every 5 years)
- Remote sensing/aerial photography (once)
- Ambient air quality and ozone injury (continuous)

## Aims of the Programme

The major objective of the 'Pan-European Programme for the Intensive Monitoring of Forest Ecosystems' is to gain a European wide overview of the impacts of air pollution and other stress

factors on forest ecosystems. An overview of the most relevant relationships to be derived with the data in the Intensive Monitoring database is given in Fig. 1.1. The results should be useful for the evaluation of (protocols on) air pollution control strategies used within the UN/ECE Convention of Long-Range Transboundary Air Pollution and the EC. More specific objectives in the context of air pollution are the assessment of:

- Responses of forest ecosystems to changes in air pollution by deriving trends in stress factors and ecosystem condition.
- The fate of atmospheric pollutants in the ecosystem in terms of accumulation, release and leaching.
- Critical loads and critical levels of atmospheric pollutants (SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, metals) in view of ecosystem effects in relation to present loads.
- Impacts of future scenarios of air pollution on the (chemical) ecosystem condition.

Recently, the aims of the Pan-European Programme have been widened towards the topics of biodiversity and climate change. In this context, the Programme aims to contribute to the development and monitoring of 'criteria and indicators for sustainable forest management' (Min conference, III.). Objectives of the Pan-European Programme related to this topic can be formulated as:

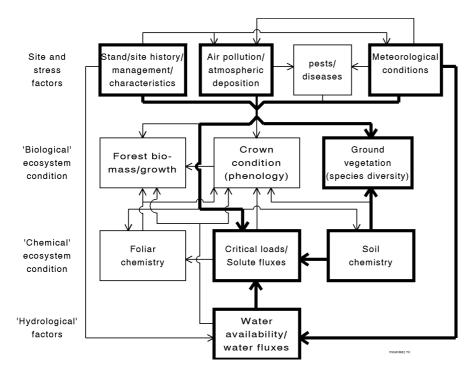
- Assessment of net carbon sequestration in European forests, to improve the assessment of the global carbon balance and to evaluate the influence of changes in the climate due to atmospheric greenhouse gasses on the forest ecosystem.
- Further development and monitoring of indicators related to the various functions of forest ecosystems to assess its long-term sustainability, such as forest ecosystem health, forest production, species composition of ground vegetation and protective functions of soil and water resources.

## 1.2 Aim of the report

The contents of Technical Reports on the 'Pan-European Programme for the Intensive Monitoring of Forest Ecosystems' in Europe differ each year in view of the increased data availability in time. In the year 1997, the first Technical Report was written containing information on the data received until 1994. This report did not contain results obtained from a data evaluation. In the period 1998-2000, three Technical reports were published of a similar character. Those reports contained information on all the surveys carried out until 1995-1997, respectively (crown, soil, foliage, increment, atmospheric deposition, meteorology and soil solution), describing the results of the different surveys, partly in relation to each other, while using statistical techniques. Since 2001, we follow another publication strategy for the period 2001-2005 (See De Vries et al., 2001) follows from the strategy for Intensive Monitoring for that period (De Vries, 2000). It aims to ensure an adequate supply of policy relevant information for the coming period and an alternation of a focus on abiotic and biotic aspects.

Aspects that have been investigated in this year's report are illustrated in Figure 1.1. The focus of this year's report is on:

- Relationships between both the occurrence probability of individual species and the species composition of ground vegetation and environmental factors and
- Critical loads for nitrogen, acidity and heavy metals for the forest ecosystem in comparison to element inputs from the atmosphere.



*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.* 

## **1.3 Contents of the report**

Chapter 2 provides information on the current implementation of the Intensive Monitoring Programme, including information on the selected plots in the various surveys and the submitted data and information until 1999. Results on the species diversity of ground vegetation at approximately 670 plots are presented in chapter 3. This includes a basic data analysis on numerical values and geographical patterns of species numbers and species diversity. The chapter focuses, however, on relationships between the species composition of ground vegetation and environmental factors. A division is made between the occurrence probability of individual species and the species composition of ground vegetation. Included environmental factors are "traditional factors", such as tree species, soil factors related to acidity/ nutrient availability and climate, and atmospheric deposition.

The assessment of critical loads for nitrogen and acidity in comparison to present loads is presented in Chapter 4, whereas Chapter 5 contains similar results for heavy metals. Chapter 4 presents methods that are needed to both calculate steady-state critical loads and present deposition thresholds. The latter thresholds equal deposition levels that lead to concentrations of nitrogen and acidity in soil solution that are equal to critical limits at present (not in a steady-state situation). Results obtained include critical loads and present deposition thresholds in comparison to present loads, distinguishing between impacts on the soil, ground vegetation and trees. Chapter 5 is limited to the calculation of critical loads, focusing on cadmium, copper, lead and zinc.

Chapter 6 contains a discussion, focusing on the reliability of the results on the species composition of ground vegetation and its relation with environmental factors and the critical loads for nitrogen, acidity and heavy metals. This chapter also contains overall conclusions related to the results in the previous chapters.

# 2 Programme

The Intensive Monitoring Programme is carried out on plots that were selected in such a way that it includes the major combinations of tree species and soil type in a country. In this chapter an overview of plots in the various surveys (Section 2.1) and of the data that have been stored until 1999 (Section 2.2) are presented.

## 2.1 Selected plots in the various surveys

The Intensive Monitoring Programme now includes 866 plots from 30 participating countries. Some countries that participate in the ICP Forests programme, have indicated their participation in the Intensive Monitoring programme, but have not yet sent the general plot information. The number of plots that have presently been installed equals 791 of the 866 plots.

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, forest growth, 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 forest growth). According to the information received, atmospheric deposition is carried out at 499 plots. Surveys with respect to meteorology and soil solution measurements are carried out at 202 and 243 plots respectively. Furthermore, it can be concluded that ground vegetation surveys will be carried out at 632 plots, whereas the application of aerial photography is foreseen at 155 plots (Table 2.1). Several countries also plan to or do carry out additional surveys on the plots, such as phytopathology, litterfall, phenology, mycorrhizae and/or fungi and other in-depth studies to soil water regimes, gas exchange and air quality measurements.

An overview of the surveys carried out at the different plots is given in Fig. 2.1. This map is based on information submitted until February 2002 and includes data up to the end of 1999. The map makes a distinction between plots where:

- Only mandatory core surveys (crown condition, soil, foliage and increment) are carried out.
- All surveys are carried out, including the core surveys and the optional surveys deposition, meteorology, soil solution chemistry and ground vegetation
- Core surveys are carried out in combination with one or more optional surveys (mostly deposition and ground vegetation).

The map shows that number of plots at which all surveys are carried out occur mainly in a northsouth transect going from Scandinavia over Germany to France and Spain. It also shows that atmospheric deposition (at least bulk deposition, but mostly also including throughfall) is measured at much more plots than those where all surveys, including meteorological measurements and soil solution chemistry, are carried out. This includes also a west-east transect going from the UK to Poland/Hungary.

survey	s and the	e remaini		eys are o	ptional).							
Countries	Total	Crown	Soil	Foliar	Growth	Atm.	Meteo	Soil	Ground	Rem.	Air	Phen
						Dep.		sol.	Veget.	Sens.	Quality	ology
EU countries												
Austria	20	20	20	20	20	20	2	2	20	20	-	-
Belgium Flanders	12	12	12	12	12	6	2	6	12	-	1	-
Belgium Wallonia	9	9	9	9	9	4	4	2	8	-	-	-
Denmark <sup>1)</sup>	16	16	15	16	16	8	3	7	16	-	3	-
Germany	89	89	89	89	89	86	66	78	80	49	60	15
Greece	4	4	4	4	3	4	4	-	4	-	4	4
Spain	53	53	53	53	53	12	12	6	52	-	12	12
France <sup>2)</sup>	100	94	100	94	94	24	25	14	94	14	24	-
Ireland <sup>3)</sup>	15	15	15	15	15	3	8	3	9	15	-	-
Italy <sup>4)</sup>	25	25	25	25	25	17	15	2	25	20	25	25
Luxembourg	2	2	2	2	2	1	2	-	2	-	-	-
Netherlands	14	14	14	14	14	7?	-	7'	14	-	-	-
Portugal	9	9	9	9	9	1	1	1	9	-	-	-
Portugal Azores	4	4	4	4	4	1	1	1	-	-	-	-
Finland	31	31	31	31	31	16	12	16	31	-	-	3
Sweden	100	100	100	100	100	46	-	46	98	12	-	-
United Kingdom	10	10	10	10	10	10	2	7	10	-	-	-
Total EU	510	504	509	504	503	263	159	200	485	130	128	59
Non-EU countries												
Bulgaria	3 <sup>5)</sup>	3	3	3	3	3	3	3	3	?	-	-
Belarus	81 <sup>5)</sup>	81	81	81	81	-	-	-	-	-	-	-
Switzerland	16	16	16	16	16	13	16	7	16	16	-	-
Czech Republic	14	14	14	14	14	5	2	3	11	-	5	-
Estonia	6	6	6	6	6	5	-	2	6	-	-	-
Croatia	7	7	7	7	7	2	3	3	4	?	-	-
Hungary	14	14	14	14	14	14	14	-	14	-	-	-
Lithuania	9	9	9	9	9	-	-	-	9	9	-	-
Latvia	2	2	2	2	2	2	2	2	2	-	-	-
Norway	19	19	19	19	19	19	-	19	19	-	-	-
Poland	148	148	148	148	148	148	-	-	148	?	-	-
Romania	13	13	13	8	13	4	-	4	13	?	-	-
Russia	12	12	12	12	12	12	-	-	-	?	-	-
Slovenia	35)	3	3	3	3	2	3	-	-	?	-	-
Slovak Republic	9	9	9	9	9	7	-	-	-	2	-	-
Total non-EU	356	356	356	351	356	236	43	43	245	27	5	0
Total	866	860	865	855	859	499	202	243	730	157	133	59
$\frac{1}{1}$ A plata raplaced of			000	555	307	.,,	202			101	100	57

 Table 2.1
 Overview of the number of selected plots for the main surveys (Crown, Soil, Foliar and Growth are core surveys and the remaining surveys are optional).

<sup>1)</sup> 4 plots replaced after storm 1999

<sup>2)</sup> 4 plots destroyed, 4 plots damaged, but all still part of network

 $^{3)}$  2 plots windblown

<sup>4)</sup> Selected plots: 27

<sup>5)</sup> In these countries plots have not yet been installed.

## 2.2 Submitted data and information until 1999

Table 2.2 gives an overview of the number of installed plots, and the number of plots for which data, DAR-Q and both data and DAR-Q's are stored. Table 2.2 shows that for the vast majority of the plots with stored data, also the DAR-Q information is available. This table furthermore shows that the number of plots for which both data and DAR-Q information were stored is (slightly) lower than the number of installed plots. The main reasons for these differences are:

- Some countries have not submitted data for some of the surveys.

- Some countries submitted data that were not stored because the data were incomplete or problems exist 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.

Inversely, the number of plots for which data are stored is generally larger than the number where assessments are presently carried out, since some plots have been abandoned (Compare Table 2.1 and 2.2).

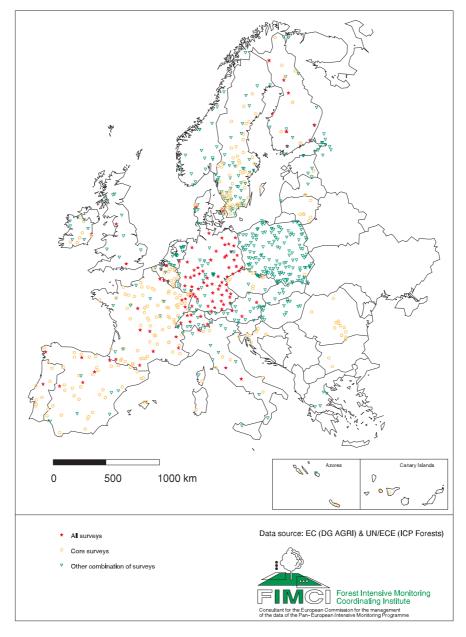


Figure 2.1 Geographical distribution of installed Intensive Monitoring plots based on information received until February 2002. Core surveys include crown condition, soil, foliage and increment, whereas all surveys include the core surveys and deposition, meteorology, soil solution chemistry and ground vegetation

Survey	Selected	plots <sup>2)</sup>	Data stored		DAR-Q information		Data and DAR-Q	
					stored		informati	on stored
	EU	non-EU	EU	non-EU	EU	non-EU	EU	non-EU
Crown condition	504	356	509	256	507	244	501	231
Soil condition	509	356	496	198	446	234	440	192
Foliar condition	504	351	513	238	453	244	448	229
Growth	503	356	492	91	472	94	452	65
Deposition	263	236	298	217	259	157	294	180
Meteorology	159	43	151	26	168	28	147	24
Soil solution	200	43	211	31	201	29	195	29
Ground vegetation	387	245	465	209	348	214	323	194

*Table 2.2 Overview of the number of plots for which data and/or information was submitted for the eight surveys until the year 1999*<sup>1)</sup>.

<sup>1)</sup> For soil, foliage and increment, also data from earlier years have been used.

<sup>2)</sup> The number of plots for which data are stored is sometimes higher than the number of plots selected, because at a number of plots measurements were stopped in the last years: for these plots only short datasets of the period before 1998/1999 are available.

Compared to last years' report, the number of plots with data has only slightly increased for most surveys. The largest increase is again found for ground vegetation as a number of countries have submitted both data and DAR-Q information last year. Ground vegetation data are now available for more than 85% of all plots, ground vegetation DAR-Q 's for about 70% of the plots.

# 3 The vascular species composition of ground vegetation

## **3.1 Introduction**

## Concern on forest biodiversity

The concern about forest decline in the 1980s lead to the initiation of nation-wide research programs, which mainly focused on the relation between atmospheric deposition and tree health or tree growth. The strong research effort in these programs yielded many new insights into the ways in which atmosphere, soil and vegetation interact. Later, forest dieback appeared to be a rather localised phenomenon, although large scale effects on forest due to multiple stress have been found. At the same time the atmospheric concentrations of sulphur dioxide strongly decreased all over Europe, and the fear for large-scale forest dieback decreased concomitantly. After the Rio convention (Agenda 21, 1992), however, there was a growing concern over the world-wide loss of biodiversity. In the text of the Convention, biodiversity is defined as 'the variability among living organisms from all sources, inter alia, terrestrial, marine and other aquatic ecosystems and the ecological complexes of which they are part; this includes diversity within species, between species and of ecosystems'. One aspect of forest biodiversity is the plant species composition of the ground vegetation of forests. In many countries a shift in species composition is reported (Ellenberg, 1985; Tyler, 1987; Thimonier et al., 1992; Van Dobben et al., 1999). This shift usually entails a transition from a cryptogam-dominated to a grass-dominated undergrowth, and an increase in species indicative for nitrogen-rich circumstances. Both acidification and nitrogen enrichment, in response to atmospheric deposition of sulphur and nitrogenous compounds, are now accepted as factors that negatively affect the diversity of plant species in forests (e.g. Bobbink et al., 1998). Furthermore, grazing of (large) herbivores do affect plant species composition (e.g. Ellenberg, 1988).

## Ad hoc Working Group on Biodiversity: Assessments of forest biodiversity parameters

In the Expert Panel meeting on Ground Vegetation (September 2000, Lillehammer, Norway), the topic of biodiversity was brought up and an ad hoc Working Group on Biodiversity was convened in Dublin (11/12 January 2001). The purpose of this ad hoc working group is to determine the possibility and feasibility of including aspects of biodiversity assessments into the current monitoring programme by:

- Investigating the possibility to include aspects of biodiversity in forests in the Pan-European monitoring programme;
- Formulating in general wordings how to assess biodiversity in forests using available data;
- Developing methods to determine specific aspects of biodiversity in forests, e.g. numerical indices of ecological value;
- Setting up a manual describing methods to harmonise aspects of biodiversity within the Pan-European Programme.

Although the monitoring programme has been designed and implemented to assess the effect of atmospheric deposition on forest condition, it can also be used to address the topic of biodiversity in forests. The monitoring programme can contribute to aspects of biodiversity assessment with information on the species composition of the ground vegetation, crown condition data (with resulting information on tree species) and tree structure (growth survey). At the intensively

monitored plots, information is also collected on environmental aspects such as soil and soil solution chemistry, atmospheric deposition and meteorological conditions. These factors may have an effect on the biodiversity parameters and as such they are also relevant. Examples are:

- Stand and site characteristics, such as soil type, stand age, altitude, tree height and diameter at breast height, and stand history (available at part of the plots only).
- Climate, such as precipitation and temperature.
- Indicators of nutrient availability, such as foliar, soil and soil solution chemistry and atmospheric deposition data.

## Aims of this study on ground vegetation composition

In this chapter we focus on the species composition of the ground vegetation, being part of the biodiversity of the forest ecosystem. The focus of this study is to relate vegetation composition to environmental conditions at a given point in time. The combination of ground vegetation data and environmental data sampled over a large part of Europe are now available and offer a unique opportunity to achieve a better understanding of the relations between the species composition of the ground vegetation and environmental factors. This is done by using statistical approaches focusing on both the individual species and on the species composition. The first approach is also relevant in parameterising models predicting the long-term impacts of environmental changes on species diversity (the SMART-MOVE model; e.g. Latour et al., 1994; Kros et al., 1995). In this way, it is be possible to identify the environmental factors that most strongly determine the geographic variation in species diversity of the ground vegetation. It specifically allows us to gain insight whether atmospheric pollution, which is considered to be one of the pressure indicators affecting species composition in forests, is one of the important factors. If such factors are known, it might be possible to assess more precisely threats to species diversity, to which local governments might anticipate. Country results related to this topic are summarised in Annex 1.

It should be stressed that this study is only based on the spatial pattern of both vegetation and predictors. It does not include temporal changes of both the species composition of the ground vegetation and environmental factors. Actually, the major aim of ground vegetation monitoring is to (i) detect temporal changes in vegetation, using vegetation as early warning signal for environmental impacts and (ii) relate those changes in vegetation to environmental changes. This aim is outside the scope of this study, since we do not yet have repeated measurements of the species composition of the ground vegetation. It may be possible that there is an effect of e.g. deposition on vegetation in time, which does not yet appear in space, but this kind of results will only become possible when sufficient repetitive measurements are available.

## Contents of this chapter

This chapter focuses on relationships between the species composition of ground vegetation and environmental factors. First, it includes information on methodological aspects (section 3.2) including basic information on the investigated plots (Section 3.2.1), data assessment methods and data comparability (Section 3.2.2, and data evaluation methods (Section 3.2.3). The data evaluation methods focus on methods to assess impacts of environmental factors on ground vegetation. It presents the used (statistical) approaches (Section 3.2.3.1) with a further distinction between the occurrence probability of individual species (Section 3.2.3.2) and the species composition at the community level (Section 3.2.3.3). The results in Section 3.3 use a similar subdivision. It starts with a basic data analysis in Sections 3.3.1 (numerical values) and 3.3.2 (geographical patterns). The results of the relation between vegetation and environmental

predictors are presented in the Sections 3.3.3 (occurrence probability of individual species) and 3.3.4 (species composition at the community level).

## **3.2 Methods**

## **3.2.1** Locations

Figure 3.1 shows the locations for which ground vegetation data up to 1999 are available. For these locations, at least data on vascular plants have been submitted. For many of the plots data on mosses and lichens are also available, but for reasons of comparability these groups have been excluded from the present analysis. The present analysis focused on the relation between ground vegetation and environmental factors, such as stand and site characteristics, climatic factors and atmospheric deposition. In this study the focus was on making use of bulk deposition data, since use of throughfall data leads to a strong reduction in the number of plots that can be investigated. Actually, the implicit assumption is that the variation in bulk deposition is comparable to the variation in total deposition. In annex 2, results of a study including throughfall and total deposition as predictors, respectively, is presented and the main results of that study are also presented in the main text. A selection of plots was used for which data on both vascular plants and these environmental factors were available. The plots used in the analysis are indicated by a red colour in Figure 3.1 (this refers to bulk deposition only).

Data on the numbers of available plots, distinguishing between all available data and the data used in the assessment of relationships between the species composition of the ground vegetation and environmental factors, are presented in Table 3.1. In total, ground vegetation data were available for 674 plots (669 plots when limiting to vascular plants), while 366 plots were used to derive relationships with environmental factors. At most plots, the availability of atmospheric deposition data was the limitation in the derivation of relationships.

Table 3.1Overview of the number of plots for which ground vegetation data were available up to 1999. Data in<br/>brackets are those used in the assessment of relationships between the species composition of the ground<br/>vegetation and environmental factors.

vegetation and en	ivironmeniai j	aciors.					
Countries	Nr of plots			Nr of plots			
	Unfenced	Fenced	All	Vascular	Bryophytes <sup>1</sup>	Lichens <sup>1</sup>	All
				plants			
Nordic countries <sup>2</sup>	165(98)	0 (0)	165 (97)	162	61	72	165
Central European countries <sup>3</sup>	401(218)	156 (72)	424 (241)	423	44	9	424
Southern European countries <sup>4</sup>	81 (24)	23 (17)	85 (28)	84	7	22	85
Total	647 (340)	179 (89)	674 (366)	669	112	103	674

<sup>1</sup> Data on bryophytes and lichens were not used in the study

<sup>2</sup> Includes Finland, Sweden, Norway, Denmark and the Baltic countries (Estonia, Latvia and Lithuania)

<sup>3</sup> Includes all countries in Europe excluding those mentioned under 2 and 4

<sup>4</sup> Includes Spain, Portugal, Italy and Greece.

## 3.2.2 Data assessment methods and data comparability

As stated above, ground vegetation data were submitted for 674 plots and on 366 of those plots bulk deposition data were also available. Information on the methods used was submitted in data assessment report questionnaires (DAR-Qs) for 553 of the plots with ground vegetation data and 307 of the plots with deposition data.

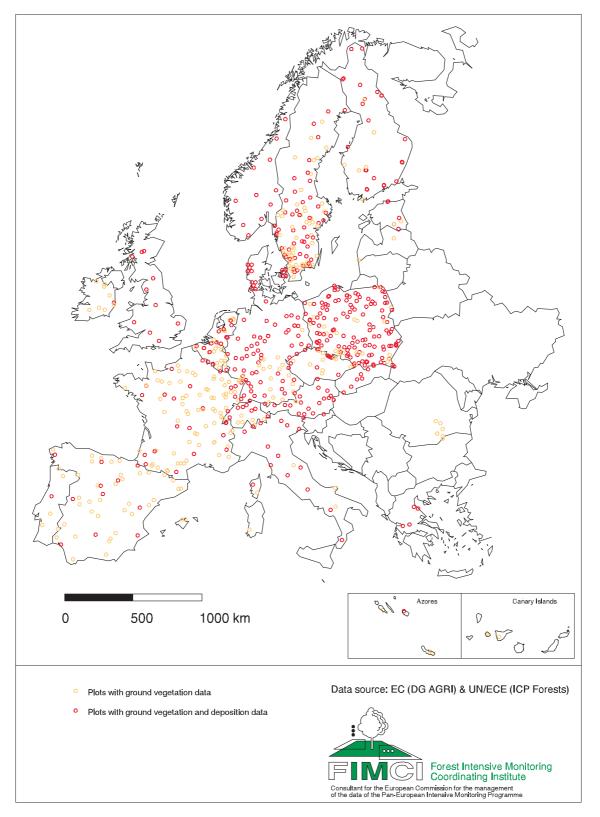


Figure 3.1 Location of the plots with available ground vegetation data up to 1999, distinguishing between the plots used for the determination of the relationship between ground vegetation and environmental factors (red) and the remaining plots.

#### General approach

The vegetation of each stand was described by means of sample plots within that stand. The species are entered in the database coded as nine-digit codes: three digits for the family name, three digits for the genus name and three digits for the species name. This standardised code was taken from the PANDORA database, which is considered as the most up-to-date checklist of the European Flora. This list can be viewed on http://www.rbge.org.uk/. When species are encountered that are not in this list, countries are allowed to make their own additions to this list. Such additions are recognisable by a different code for the latter three digits followed by a code for the country. Agreements that have been made with respect to the species codes for mosses and lichens can be viewed in: http://www.skogforsk.no/forskning/skogpatologi/ops/icp-for-veg/Code\_Lists\_Cryptogams/Default.html

In the sample plots, estimates of the abundance, i.e. the quantity of each species, were made. Estimates of species cover were always made by eye in the field, as perpendicular projection of all living parts on the ground surface. The most commonly used method was a direct percent scale approach. Some fieldworkers first made their estimates in class intervals, for which various coding scales exist, the most commonly used being Braun-Blanquet (Table 3.2). All codes were back transformed to percentages before entering the database. The information given in Table 3.1 includes all plots, but also the plots that were used to derive relationships between the species composition of the ground vegetation and environmental factors. In this case, the available data on (bulk) deposition was the major limiting factor.

Table 3.2	Number of plots in which the abundance ground vegetation is assessed directly
	or indirectly by a coding scale and a combination of both. Numbers includes all
	plots and the plots that were used to derive relationships between the species
	composition of the ground vegetation and environmental factors
Alarmalama	Number of slats

composition of the ground vegetation and environmental factors					
Abundance scale	Num	ber of plots			
	All plots	Plots for relationships			
% scale	326	201			
Braun-Blanquet	197	86			
Other <sup>1</sup>	30	20			
Data, but no DAR-Q	121	59			
All plots	674	366			

<sup>1</sup> Including Londo and combinations such Barkman et al. + %scale, Braun-Blanquet + %scale and Braun-Blanquet + Londo

Ground vegetation was assessed in various layers that have been divided on the basis of either taxonomic groups (i.e., mosses, vascular plants), morphology (e.g. shrubs, trees), height (e.g. <0.01m, 0.01-0.5 m, 0.5-10 m and >10m) or a combination (Table 3.3).

Table 3.3 Number of plots in which ground vegetation is divided by taxonomy and/or height, divided in all plots and plots that were used in deriving relationships with environmental factors.

Division in layers	Number of plots				
	All plots	Plots for relationships			
Taxonomic groups	29	17			
Height	34	26			
Taxonomy and height	465	263			
No information	25	1			
Data, but no DAR-Q	121	59			
All plots	674	366			

To circumvent the problems arising from the use of differently defined layers, no distinction between layers was made, and the abundance's of each species in the various layers were combined by taking the maximum abundance per species.

The number and size of the sample plots and the methods of quantitative assessment for each species (most often in abundance classes) differ regionally. In a given stand, the number of species observed in sample plots a priori depends on (i) total sample area of the plot, including position and size of the sampling units, and (ii) the measurement of species inside or outside a fence. These differences are described in more detail below.

## Sample area and fencing

In determining the species composition of the ground vegetation, many countries used a number of sampling units within the plot. The information on sub-plot level, however, has to be combined to plot-level before submission of the data. Information on subplots is therefore not available at FIMCI. Table 3.4 presents an overview of the total sampled area used in the 674 plots for which ground vegetation data were submitted and the 366 plots used in deriving relationships with environmental factors.

Table 3.4Total sampled area of each plot.

Total sampled area (m <sup>2</sup> )	Number of plots	
	All plots	Plots for relationships
10-50	72	51
51-100	179	128
101-400	74	54
401-1000	187	53
1001-2500	41	21
Data, but no DAR-Q	121	59
All plots	674	366

The total sampled area varied between 10 and 2500 m<sup>2</sup>. In order to allow comparison of the data, normalisation of the number of species to a standard area (e.g. 400 m<sup>2</sup>) might be needed. This aspect has been dealt with in the previous Technical Report, by relating the species number to sample area, climate zone and number of trees per hectare (De Vries et al., 2001). This analyses showed no statistically significant relationship between plot area and species number. Therefore the total sampled area was not used as a predictor variable in our analyses. From the literature, the impact of plot size on species numbers is, however, well reported and the lack of a result might be caused by all other differences between plots apart from plot size. In principle, such a study should therefore be carried out by comparing the species numbers in plots of different areas within the same plot, but such data were not available.

Fencing may have a strong effect on the vegetation within a relatively short time (1-5 years) due to the exclusion of grazing (Kuiters and Slim, 2000). The present data contain both fenced and unfenced plots (Table 3.5) and therefore allow the determination of the effects of this measure. In last years report (De Vries et al, 2001) a comparison was made between the number of species in fenced and unfenced plots at 36 plots where ground vegetation was assessed at the same day and with the same sample area. Those results did not yet show significant differences. This year, an additional test was carried out to determine the effect of fencing, by splitting the plots in a fenced and an unfenced part (if applicable) en treating 'fence' as an extra class predictor. In an analysis that used all available plots (including the ones with deficient deposition data) the effect of this predictor also proved to be non-significant (even at p=0.1). The paired comparison of fenced and

unfenced plots, as done before, is much more sensitive but such a comparison was only possible for few countries using paired fenced and unfenced plots and this lead to the same conclusion. As most plots have been installed only a few years ago, it may at this date still be too early for a significant effect. Therefore the fenced and unfenced plots were taken together and treated as a single plot for the multivariate analysis and the analysis of species numbers and species diversity indicators (Simpson index).

Table 3.5	Number of fenced and unfenced plots and a combination of both, divided in all
	plots and plots that were used in deriving relationships with environmental factors.

Type of plot	Number of plots	
	All plots	Plots for relationships
Unfenced	362	218
Both unfenced and fenced	175	75
Fenced	9	9
No information	7	5
Data, but no DAR-Q	121	59
All plots	674	366

## Data comparability

In evaluating the data, it is essential to know whether the methods used in different countries create a problem for a common European evaluation of the species composition of the ground vegetation and whether the quality and accuracy of the observations are sufficient. In performing the analyses, the following assumptions were made:

- The inclusion of only vascular plant gives an adequate description of species composition of the ground vegetation. The impact of excluding bryophytes and lichens on the species composition assessment is thus implicitly assumed to be small. The consequence of this point is further discussed in Section 3.4.3
- Differences in the sampling programme, in view of the size and design of the sampled area and use of fencing, which do affect the results on the species composition of the ground vegetation, were preliminary included by using country as a predictor variable. Since "country" includes not only different methods but also different ecological circumstances, besides factors like climate and soil that we do include in our analysis, this approach only gives a rough impression of the possible impact of methodological differences between countries. In the future, the impact of real methodological differences should be assessed, based on e.g. field comparison by assessment teams.
- By using the species number and the Simpson diversity index (see Section 3.2.3.1), we characterise species composition in such a way that the results are comparable on a European scale.

These aspects, including an evaluation of the need for a revised manual on data assessment methods and a quality assurance programme, are discussed in more detail in Section 3.4.3.

## **3.2.3 Data evaluation methods**

## **3.2.3.1** General statistical approach

## Univariate and multivariate analysis

Many models that are applied to relate ecological effects to variations in single environmental variables are functionally mechanistic of nature. It is considered questionable (Latour et al., 1994)

whether mechanistic modelling can predict the combined effects of a multitude of environmental variables. As an alternative, the magnitude of these effects may be estimated by applying a statistical approach. However, for statistical models the availability of a comprehensive data set is a prerequisite. Since this is the case with the ground vegetation data at the Intensive Monitoring plots, this approach was used in the present study.

The statistical analysis of ecological datasets is often complicated by the large number of dependent variables, namely the species. In the present data there are 2121 species. Two approaches are possible to overcome this problem: an automated species-by-species univariate analysis, or a multivariate analysis. In the latter case, mutual correlation between the species are determined so as to identify the most important directions of variation in the species data, which are subsequently or simultaneously related to environmental predictors. As both methods have their pros and cons, both approaches have been applied to the present data.

The univariate statistical analyses have been carried out by the program packages S-Plus 2000 Professional Release 3 (Data Analysis Products Division, MathSoft, Seattle, WA; analysis of individual species), and GENSTAT (Payne et al., 1993) (analyses of species numbers and Simpson index), while the graphical representations of the species responses have been prepared with the Excel-97 plug-in Crystal Ball (Decisioneering Inc., Denver CO). The multivariate analyses was carried out with CANOCO (Ter Braak and Smilauer, 1998) and the biplots have been prepared with the program CANODRAW (Smilauer, unpublished).

## **Response and predictor variables**

In the species-by-species univariate analysis, the occurrence of a species (which can take the values 1 to indicate its presence and 0 to indicate its absence) is the response variable. In the multivariate analysis, the abundance values of all species together form a multidimensional response variable. In this analysis, the log number of species and the log Simpson index have been used as the response variables. The number of species was used because it is the most straightforward measure of plant species diversity, which however cannot be directly entered into statistical analysis because of its very skewed distribution (Figure 3.5). The Simpson index was calculated according to:

$$D = 1 - \sum_{i=1}^{n} p_i^2$$
(3.1)

in which  $p_i$  is the cover fraction (percentage divided by 100) of species i in the plot. In applying this equation the summed percentage cover of all species in a plot was normalised to 100%. D varies between 0 and 1, a higher value indicating the presence of many species in approximately equal quantities. An advantage of Simpson's D over other diversity measures is its clear ecological interpretation, namely as the probability that two individuals picked at random from the community, belong to different species. The philosophy behind biodiversity indicators has already been treated in the previous Technical Report (De Vries et al., 2001). When interpreting the results it should be kept in mind that the two biodiversity measured used are rather strongly correlated in the present data (r=0.57, n=669).

The predictor variables that were used in this study in both types of analyses is given in Table 3.6. Variables related to forest management were not included, because this kind of information was grossly lacking. Regarding deposition, use was made of bulk deposition data, since use of throughfall would lead to a strong reduction in the number of plots that could be investigated (see also Section 3.2.1).

Table 3.6 is based on both the relevance of predictors and the availability of data. In all cases, use was made of the data that were available in the Intensive Monitoring database, except for temperature which was partly derived by interpolating data from nearby meteorological stations. A possible future development would be to also consider seasonal variation in temperature and precipitation, rather than just annual precipitation and annual mean temperature, since differences in winter temperatures may be important for some species, even though annual mean temperatures were the same. Relevant predictors that were not yet available were canopy closure, slope and exposition. Also for these predictors the use of other information sources in the future should be considered. For stand age, plots that were classified as 'uneven aged' have been assigned a notional age of 65 years. The predictor variables related to soil chemical data and bulk deposition contained only few missing variables (< 5% for only a few parameters) because variables with many missing values were excluded beforehand. Missing values were replaced by a best estimate based on regression equations with other available parameters in the data set. In this way, for 12 plots, base saturation was estimated from known pH (r=0.67); for 3 plots CEC was estimated from the organic C content (r=0.4); for 1 plot NH<sub>4</sub> bulk deposition was estimated from NO<sub>3</sub> in bulk deposition (r=0.52); and for 1 plot K content was estimated from known Mg content (r=0.32). Only for Ca in the humus layer about 9% missing values occurred which were replaced by estimates using the correlation with Mg content (r=0.76).

Table 3.6 Environmental predictors used in the statistical analysis of ground vegetation data
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Predictor variables	
Stand and site characteristics	
- Country (categorical)	

- Climate zone (categorical)
- Soil type (categorical)
- Main tree species (categorical)
- Stand age (continuous based on discrete intervals)
- Stand height (continuous based on discrete intervals)
- Altitude (continuous based on discrete intervals)
- Soil cover (calculated from number of trees and diameter at breast height

## Climatic variables

- Annual precipitation
- Annual average temperature (partly from Level II data base, otherwise interpolated values from meteorological stations)

## Air pollution influence

- Bulk deposition (throughfall or total deposition<sup>1</sup>) of NH<sub>4</sub>, NO<sub>3</sub>, SO<sub>4</sub>, Ca, Mg, K, Na and Cl *Soil chemical data related to nutrition* 

- Organic mass of humus layer
- C, N, P, Ca, Mg and K content in the humus layer (partly based on foliar data)
- C and N content in the mineral topsoil (0-20 cm)

## Soil chemical data related to acidity

- The pH (CaCl<sub>2</sub>) of the humus layer and mineral topsoil (0-20 cm)
- CEC and base saturation in the mineral topsoil (0-20 cm)

In Annex 2, results are given of an approach in which throughfall and total deposition data (of less plots) were used

## Ellenberg indicator values

Several attempts have been made to classify species according to their ecological response (e.g., Grime et al., 1988; Diekmann and Dupré, 1997; Hawkes et al., 1997). Of these classifications, the Ellenberg indicator (1991a) is most widely used. It scores the response of each species on an

arbitrary nine-point scale for seven environmental factors. This includes light availability, temperature, 'continentality' (the East-West distribution pattern), water availability, soil pH, nutrient availability and salt tolerance. For example a score with 1111111 stands for extremely dark, cold, oceanic, dry, acid, nutrient-poor, low Cl. On the water availability scale, three additional classes 10-12 are used for aquatic species. Ellenberg's database includes 2792 species with values for at least one of the indicators. Although the database was developed for Central Europe (Germany, Austria, Switzerland), its validity outside this region, specifically in Northern and Western Europe, has been shown by several authors (e.g., Thompson et al., 1993; Hannerz and Hanell, 1997; Wamelink et al., 2002). This refers to Britain and Norway, as well as for Sweden, Denmark, Poland and parts of France. It does not hold for Southern European countries Nevertheless, the procedure is applicable for most of the plots included, since most data refer to the Central and Northern European part.

To use the Ellenberg response classification for the present ground vegetation data, a table was constructed that translates Ellenberg's nomenclature into the PANDORA nomenclature. This resulted in Ellenberg indicator values for 656 out of a total of 967 species (or 360 out of the 396 species with more than two occurrences) for the 366 plots that were used for the determination of the relationship between vegetation and environment, or 911 out of a total of 1535 species in the complete dataset (674 plots). For each plot Ellenberg scores were calculated as the unweighted means over all species with known Ellenberg indicator values present in the plot (excluding the trees). The resulting scores were checked for normality. Salinity scores were not used because of their extremely skew distribution caused by a unit value for most plots.

It should be stressed that the Ellenberg values in general are not based on measured species responses, but are largely based on expert knowledge which may contain considerable bias (Wamelink et al., 2002). Therefore in the present analysis the Ellenberg values were only used as an aid to the interpretation of the results, in addition to the author's expert knowledge. This means that conclusions on species composition of the ground vegetation and its relationship with the abiotic environment are not based on Ellenberg values, however at several points it was checked whether the observed responses of the species to their environment was plausible on the basis of their Ellenberg values.

## Data treatment

Table 3.7 summarises the treatment of the species data. The tree layer was removed from the data because the analysis of the undergrowth vegetation was the objective of this study. Instead, the main tree species was used as a predictor variable, but the tree species still have an influence on the vegetation level because seedlings of the species in the tree layer also occur in other layers. The 'local' species (i.e., the ones added by the individual countries to the standard species list) were also removed because of their unclear taxonomical status. Furthermore, in some analyses the country was used as a predictor, and these species are confounded with the countries. When the same species occurred in more than one layer, the maximum abundance over the layers was taken. A separate treatment of layers was not feasible because the layer definitions differed among the countries.

Next, the subplots within each single plot were taken together as the mean value over each species. The resulting total plot size strongly differed between (or sometimes even within) the countries. It was assumed that plot size has no effect on the results as presented here, but this assumption still remains to be checked in further analyses. A preliminary analysis (see Technical

Report 2001) showed no significant effect of plot size on total species number. If a plot was sampled more than once within the year of observation, these observations were taken together as the maximum for each species. A check was performed to verify that all observations in a plot were from the same year. In the resulting dataset, all Rubus species that belong to the R. *fruticosus* aggregate, were taken together (the reason for this was that the taxonomic concepts of *Rubus* are different in each country and PANDORA does not recognise the aggregate species *R*. fruticosus). This dataset (669 plots with >0 vascular species, 1553 species) was used to draw maps of species numbers, Simpson index and Ellenberg indicator values.

Action	Number of species	Number of subplots	GL <sub>1</sub>	GL <sub>2</sub>	$(\lambda_1+\lambda_2) / \Sigma\lambda * $ 100%
remove tree layer and local species	1543	2973	n.d	n.d.	n.d.
lump layers to maximum per species	1543	2973	n.d	n.d.	n.d.
lump subplots to mean per species	1543	877	n.d	n.d.	n.d.
lump plot observations within a year to maximum per	1543	674	n.d	n.d.	n.d.
species					
lump <i>Rubus fruticosus</i> aggregated to maximum <sup>1</sup> )	1535	674	n.d	n.d.	n.d.
select plots with abiotic data	967	366	n.d	n.d.	n.d.
remove plots with no vascular species $^{2}$ )	697	362	54.7	9.9	6.8%
remove species occurring only once	551	362	42.8	8.4	7.9%
remove one plot with very deviant species	551	361	8.0	6.5	6.8%
composition					
remove species occurring only twice	396	361	12.8	13.0	8.6%
downweight rare species	396	361	37.7	7.5	13.7%
remove one plot with outlier in species $>$ predictor	396	360	17.4	36.6	15.7%
relationship <sup>3</sup> )					
remove species occurring only three times	316	360	22.8	7.6	13.7%

Treatment of the data. GL = gradient length,  $\lambda =$  eigenvalue, subscript = axis number, n.d. = not Table 3.7 determined

<sup>1</sup>) this dataset was used to draw the maps of diversity and Ellenberg indicator values

 $\binom{2}{2}$  this dataset was used for the analysis of species numbers and Simpson index after removal of two outliers  $\binom{3}{2}$  this dataset was used for the multivariate analysis

Statistical treatments were carried out on a selection of the plots for which most of the predictor variables given in Table 3.6 were available. The resulting dataset (360 plots, 967 species) was used for the univariate analysis. For the multivariate analysis it was attempted to further reduce heterogeneity. This was done by removing rare species, and also by applying a downweighting procedure. This procedure entails a reduction of the weight of species that have a frequency of <0.2\*(frequency of the most common species), inversely proportional to their frequency. The resulting gradient lengths and relative eigenvalues (see Section 3.2.3.3) of the first two axes are given in Table 3.7. A dataset with the species with less than three occurrences removed, and a downweighting procedure as described above, resulted in a maximum in  $(\lambda_1 + \lambda_2) / \Sigma \lambda$  and a not too large gradient length for the first axis. After the removal of two outliers this dataset (360 plots, 396 species) was used for the multivariate analysis.

For the multivariate analysis and the analysis of biodiversity, all predictor variables were transformed according to  $Z' = \ln(Z-MEAN[Z]+1)$ , except pH and the class variables. The class variables were split into dummy variables (one for each class, 1 for samples that belong to that class, else 0). Histograms of the thus transformed predictors were inspected by eye, but no outliers or very skew distributions were found. Additionally, the 'leverage' (a measure for the relative influence of each sample) was used as a check on outliers (both for each individual predictor and in the full predictor + co-variable space). In canonical correspondence analysis (CCA), all (log-transformed) predictors were standardised to zero mean and unit variance before

the analysis. The species abundance's were transformed according to Y' = ln(Y+1). For the perspecies analysis, the original quantitative variables were used (not transformed), and the class variables were not used at all.

#### 3.2.3.2 Univariate multiple regression analyses of individual species

#### Generalised Linear Model approach

Multiple regression can be used to formally express the occurrence probability of individual species as a function of environmental factors and their interactions. This type of regression modelling is based on covariance of the species and environmental predictors. The required information is not necessarily quantitative; also class data (e.g., soil types or species presence/absence) can be analysed in this way.

Latour and Reiling (1993) developed a conceptual, species-centred, multiple-stress MOdel for VEgetation (MOVE), which relates the occurrence of individual species of plants to nutrient availability, pH and moisture content. In order to calibrate the MOVE model, the response curves of 700 Dutch plant species have been constructed for the combination of soil moisture content, nutrient availability and soil acidity, as estimated on the basis of Ellenberg's (1991a) indicator values (Wiertz et al., 1992). In the present study a comparable method was applied, resulting in estimates of the species' responses that can be depicted in the form of response curves. The basis of this method is a Generalised Linear Model (GLM), with the general model:

$$\ln\left(\frac{p}{1-p}\right) = a + b_1 \times \text{categorical pred}_1 + \dots + b_x \times \text{categorical pred}_x + c_1 \times \text{scalar pred}_1 + \dots + c_x \times \text{scalar pred}_x + d_1 \times \text{scalar pred}_1^2 + \dots + d_x \times \text{scalar pred}_x^2$$
(3.2)

where p is the probability of occurrence of a particular species, and a...d are regression coefficients. The categorical predictors are binary coded (1 if a sample belongs to a given class, else 0). The quality of a GLM-regression is given as the difference between the deviance (scaled error sum of squares) of the calculated model with predictors and the deviance of the null model, being defined as:

$$\ln\left(\frac{p}{1-p}\right) = a \tag{3.3}$$

This so-called explained deviance is  $\text{Chi}^2$ -distributed with the number of predictor variables as the degrees of freedom (df). The same holds for adding single terms to the model. The added explained deviance is equal to the difference in explained deviance of the model before and after addition of the term with the degrees of freedom being one. The probability of the Chi<sup>2</sup> for term additions and the overall model is equal to the probability that the explained deviance is caused by random variation. The GLM is formulated to automatically and iteratively add significant predictor terms ( $p_{\text{explained deviance}} < 0.05$ ) to the model. The regression series have been performed with step-wise addition of the categorical predictors for climate, soil type and forest type.

#### Explained variance and number of predictor variables in derived relationships

The original dataset supplied for this study were 70728 vegetation records with a subset of 18968 records on the occurrence of 1121 herbaceous plant species that are corresponding to 422 sites in 19 countries with the required physico-chemical data. Excluding species occurring at less than 5 or more then 417 sites, limited the number of species to a manageable 332. The other subsets of data on the tree, shrub and moss layers are simply not containing sufficient data for this type of analysis. As is demonstrated in Figure 3.2, it turned out that there was very little difference in explained deviance between the most complex model and the most simple model. In this context, the most complex stands for the model including all predictor variables (see table 3.6) with the exception of Na in bulk deposition, whereas the most simple model excludes, climate, soil type, forest type and Na in bulk deposition. In both cases, the deviance mostly ranges between 10 and 70% with a median deviance around 30%.

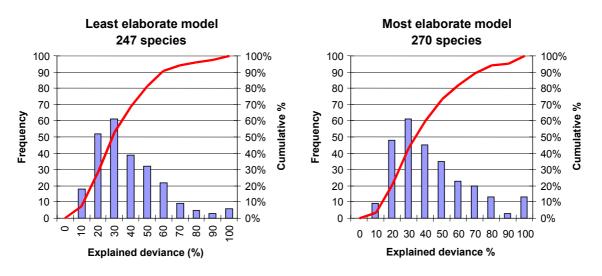
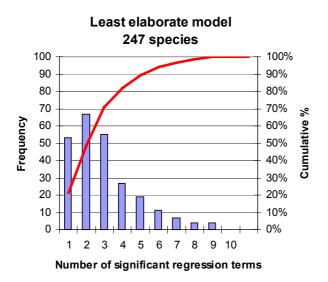


Figure 3.2 Illustration of the very minute difference in explained deviance between the most and the least complex regression models, expressed as (cumulative) frequency distributions of explained deviance over species.

The small difference in the deviance between the simple and complex model may be explained by the fact that the variability in the categorical climate predictors is also present in the scalar deposition and temperature predictors, with which they are strongly correlated. Similarly, the soil type category may largely be replaced by the soil chemistry scalars. Forest type is considered of most importance for the amount of solar irradiation received by the herb layer on the forest floor, which is also represented by the scalars on soil cover, forest age and tree height. Since the difference in explanatory power of models with different complexity is extremely small, further analysis has been limited to the least complex model.

The least complex regression formula is capable of calculating significant models (p << 0.05) for 247 species out of the original 333 species. The refusal of the statistical program to calculate a valid model for the remaining 85 species is due to a lack of covariance between the occurrence of the species and any of the predictor variables. Figure 3.3 gives the frequency distribution of the number of significant predictors for the 247 species producing a valid regression.



*Figure 3.3 Distribution of the number of explaining factors over species.* 

## 3.2.3.3 Multivariate correspondence analysis of species composition

## Principles of correspondence analysis

In multivariate analysis, the samples should be envisaged as points in a multidimensional hyperspace, their co-ordinates being given by the abundance's of the species. The analysis attempts to shift or rotate the original axes determined by the species in such a way that the most important variation in the data is represented in only a few dimensions. In doing so, the aim can be to optimally represent the relations between the species themselves (the so-called 'indirect gradient analysis'), or the relation between the species and environmental predictors ('direct gradient analysis'). Furthermore, the type of response of the species to their environment has to be considered. In a heterogeneous data set like the present one, the response of the species should be considered unimodal, i.e. each species occurs optimally at a certain point along an environmental gradient, and decreases in both directions away from the optimum. Therefore, techniques related to linear regression cannot be directly applied here because they assume a monotonous response.

In order to deal with the heterogeneity in the data, correspondence analysis (CA) related techniques were used, both in its 'indirect' ('true' CA) and in its 'direct' ('canonical' CA = CCA) form. The aim of CA is to order the samples along a notional environmental gradient, and concomitantly, order the species as to their optima along this gradient. For example, if pH is an important factor for the species composition of the samples, the species are ordered as to increasing pH optimum. Note that this can even be done if the pH itself is unknown, by rearranging the species X samples matrix so as to resemble a two-way Petrie matrix (i.e. with the nonzero values along a diagonal line; this is the classical phytosociological approach as described by Braun-Blanquet 1964). If the species are thus arranged, the samples can be ordered on the basis of their constituent species. Next, the hypothesis that pH is really causing this gradient can be tested by regressing the sample order on the measured pH. This is the 'indirect' procedure. If the environmental factors causing the gradient are known or suspected beforehand, a 'direct' procedure will usually show more clearly the relation between the species and these factors. In

the direct procedure, the regression of the sample scores on the environmental predictors is incorporated in the iterative algorithm instead of being applied afterwards (Jongman et al. 1995).

The principle of (C)CA is a repeated ordering of samples and species as described above; Table 3.8 gives a formal description of its algorithm. The axes resulting from (C)CA are always ordered as to decreasing importance, their eigenvalue  $\lambda$  being a measure of their importance. The measure  $\lambda_n / \Sigma \lambda$  can be interpreted as the fraction explained variance per axis. In direct techniques, the number of 'canonical' axes (i.e., determined by the predictors) equals the number of predictors, whereas the higher axes are determined by the species only. In this case, the sum of the canonical eigenvalues divided by the sum of all eigenvalues ( $\Sigma \lambda_{can} / \Sigma \lambda$ ) is the fraction of the variance in the species data explained by the environmental predictors.

Table 3.8The (C)CA algorithm. Explanation of symbols: u = species score, x = sample score, y = species<br/>abundance, z = predictor or covariable value, c = regression coefficient, d = centroid, s = dispersion;<br/>explanation of subscripts: i =sample, k = species, q = predictor or covariable, + = sum e.g.  $y_{+1} =$  sum of<br/>the abundances of all species in sample i,  $y_{++} =$  overall sum of abundances. Upon convergence, s<br/>becomes the eigenvalue. In CA, steps 4 and 5 are skipped (but step 12 can still be used); for the first axis,<br/>step 6 is skipped; without covariables in the analysis, step 11 is skipped. After Jongman et al. (1995),<br/>modified.

	mougreu
step	action
1.	assign random, but unequal, initial scores x <sub>i</sub> to the samples
2.	calculate new species scores as weighted sample scores: $u_k = \sum_i y_k i x_i / \sum_i y_{ki}$
3.	calculate new sample scores as weighted species scores: $x_i = \sum_k y_k i u_k / \sum_i y_{ki}$
4.	perform a multiple regression of the sample scores on the predictors using the regression equation $x=c_0+\Sigma_q c_q z_q$
5.	replace the sample scores by their fitted values from step 4: $x_i=c_0+\sum_{a}c_{a}z_{ai}$
6.	orthoginalise the axis; i.e. regress the sample scores on those of the next higher axis, and replace the original scores
	by the residual of this regression; repeat this process for each subsequent higher axis
7.	calculate the centroid d of the site scores: $d = \sum_i y_{+i} x_i / y_{++}$
8.	calculate the dispersion s (weighted variance) of the site scores: $s^2 = \sum_{iy+i} (x_i - d)^2 / y_{++}$
9.	standardise the site scores by replacing $x_i$ by $(x_i-d) / s$
10.	on convergence go to step 1 for the next axis; else go to step 2
11.	regress the final sample scores on the covariables, and replace the original scores by the residual of this regression
12.	for the construction of the biplot, calculate the correlation coefficient between the sample scores x on each axis, and
	the predictors z.

## Selection of the most influential predictors

In order to determine the relative importance of a set of predictors, a procedure of forward selection was used. In this procedure, terms leading to the highest increase in explained variance are consecutively added to the model. Two measures are used to indicate the importance of each term, namely the increase in explained variance at the moment of its inclusion in the model, and the F-value, i.e. (regression sum of squares with this term - regression sum of squares without this term) / error mean square. As in multivariate statistics no theory exists to analytically derive P-values, these are derived by a bootstrap procedure. This is achieved by a repeated random shuffling of the predictor values, and determination of the resulting F-values. Under the null hypothesis, the F-value of the data is just a random sample of the population of all possible F-values as estimated from the bootstrap samples. Or in other words: the probability of a given or higher F-value under the null hypothesis can be estimated as the order of this particular value in the population of bootstrapped F-values. In interpreting the P-values it should be borne in mind that these are only estimates that have a certain spread around a mean value; repeating the bootstrap procedure with a different randomisation may lead to slightly different estimates of P.

## Presentation in biplots

The results of multivariate analyses are usually depicted in ' biplots', which visualise the mutual relations between species, samples and predictors. There are a few simple rules for their interpretation, which are slightly different for different types of plots. For the plots as used here, the most important rules are as follows:

- There are three parts to each plot, containing information on species, samples and predictors, respectively. These should be projected over each other in equal scaling. The sample plot itself is not given here, but its data are used to determine the relation between the axes and Ellenberg indicator values (Table 3.16).
- Species are denoted by abbreviated names. The further two species are removed from each other, the lower their correlation coefficient (i.e., species located at opposite sides of the origin are strongly negatively correlated).
- Quantitative predictors are denoted by arrows. The cosine of the enclosed angle is an estimate of their mutual correlation. The projection of a species point on a predictor arrow is an estimate of that species' optimum relative to that predictor, with scaling: origin = mean, head of arrow = mean plus one standard deviation, mirror image relative to origin = mean minus one standard deviation.
- Class predictors are denoted by triangles (which in fact are the centroids of the sample scores of the samples that belong to that class). A species optimally occurs in a class if its name coincides with the triangle representing that class, and has less affinity to a class the further its name is removed from that class' triangle.

## Reduction of heterogeneity on the basis of gradient lengths and relative eigenvalues

As may be deduced from the CA algorithm in Table 3.8, very heterogeneous datasets containing many rare species lead to unstable CA solutions. Figures 3.4 and 3.5 give an impression of the heterogeneity of the present data set.

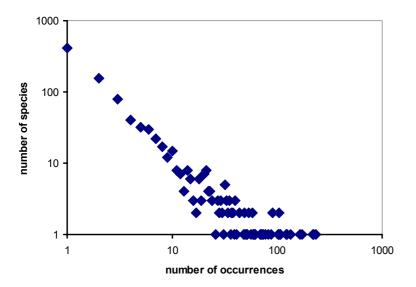


Figure 3.4 scatter diagram of number of species against number of occurrences, on a log-log scale

Of the 967 species, 651 occur in less than 1% of the plots; the average number of species per plot is 22.1, but ranges from 1 to 127. It was attempted to remove some of the heterogeneity from the data before applying CA. In doing so, two measures of heterogeneity have been used: the

'gradient length' and the sum of the first two eigenvalues relative to the sum of all eigenvalues. These measures are explained below.

The gradient length is a measure of the species turnover when moving along a given axis. It is determined by expressing the length of an axis in terms of the weighted mean standard deviation over the species. If a certain axis just represents the full Gaussian response of most species (i.e., when moving along this axis the species increase from near zero to maximum, and back to near zero again), the expected gradient length is c. 4 (because in the Gaussian model most of the observations fall within mean  $\pm 2$  S.D.). In a heterogeneous dataset, at least some of the axes represent a far longer gradient than for each single species (i.e., when moving along this axis species are consecutively replacing each other). Datasets with gradient length is < 20 may be considered very heterogeneous. On the other hand, when the gradient length is < c. 3, the response of the species can be considered linear, and other techniques than CA (i.e., the PCA-related ones) are more appropriate for the analysis of such data.

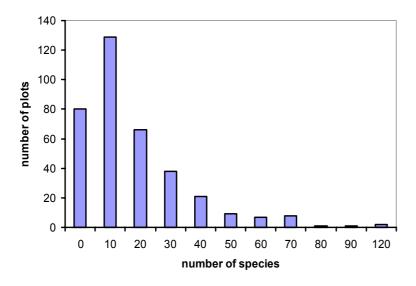


Figure 3.5 Histogram of number of species per plot

In CA, the number of axes and hence the number of eigenvalues equals the number of species. If the species are just randomly distributed over the samples, it will not be possible to reduce the dimensionality of the data, and all eigenvalues will be equal (namely,  $\Sigma\lambda$  / number of species). On the other hand, if all species are perfectly correlated, there is only one dimension in the data, and all eigenvalues will be zero except the first one. In practise the situation is always in between these extremes, and the measure  $(\lambda_1 + \lambda_2) / \Sigma\lambda$  gives an indication of the dimensionality of the data, with extremes: 2 / number of species (extremely high dimensionality), and 1 (only one or two dimensions).

#### Relation between species numbers and Simpson index and environmental predictors

It was assumed that the species numbers and Simpson index are less sensitive indicators of the species' response to the environment than the CCA-scores. The reason for this is that, if one species is replaced by another one, this will be reflected in the CCA analysis but not in these indicators of plant diversity. Therefore the statistical models derived by CCA were used as a starting point to derive models for species diversity in response to environmental factors. Using backward selection these models were simplified stepwise, until a 'minimal' model resulted, that

contained terms with a significant effect only. It was also assumed that the species numbers and Simpson index are less influenced by methodological differences, and therefore the effect of the countries was not accounted for in this analysis. As an extra check on the effect of deposition terms, these terms were added to the thus derived 'minimal' models, and the resulting models were simplified again by backward selection.

# **3.3 Results**

## 3.3.1 Ranges and correlations in response and predictor variables

Relations between species composition of the ground vegetation and the environmental factors were derived with a subset of the total available number of plots. In Table 3.9 the ranges in predictor variables are given for those plots. The relationships derived cannot be used for predictions outside this range.

Variable <sup>1</sup>	Unit	5%	50%	95%	
Response variables					
Species number	-	7	24	80	
Simpson index	-	0.156	0.677	0.908	
Stand characteristics <sup>1</sup>					
Stand age	yr	30	50	130	
Stand height	m	9.6	17	31	
Soil cover	(%)	50	70	86	
Altitude	m	25	175	1125	
Meteorology					
Precipitation	mm.yr <sup>-1</sup>	395	719	1456	
Temperature	°C	2	8	13	
Deposition					
NH <sub>4</sub>	mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup>	66	398	887	
NO <sub>3</sub>	mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup>	82	272	562	
SO <sub>4</sub>	mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup>	123	432	1190	
Ca	mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup>	37	260	932	
Mg	mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup>	26	101	455	
K	mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup>	20	61	310	
Na	mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup>	43	188	1151	
Cl	mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup>	46	257	1390	
Element contents in humus layer					
C	g.kg <sup>-1</sup>	131	325	484	
Ν	$g.kg^{-1}$	4.6	12	19	
Р	g.kg <sup>-1</sup>	0.18	0.65	1.1	
K	g.kg <sup>-1</sup>	0.41	0.96	4.3	
Ca	g.kg <sup>-1</sup>	1.1	2.5	9.5	
Mg	g.kg <sup>-1</sup>	0.19	0.59	4.2	
Element contents in mineral layer					
C	$g.kg^{-1}$	5.5	22.1	90	
N	g.kg <sup>-1</sup>	0.35	1.2	5.06	
Soil acidity aspects					
pH-CaCl <sub>2</sub> humus layer	-	2.7	3.2	5.2	
pH-CaCl <sub>2</sub> mineral layer	-	3.2	3.8	5.1	
CEC mineral layer	mmol <sub>c</sub> .kg <sup>-1</sup>	11	37	240	
Base saturation mineral layer	(%)	4.5	16.4	91.1	

Table 3.9Ranges (5 - 50 - 95 percentile) of response variables (species number and Simpson index) and predictor<br/>variables (environmental factors) used in the regression analyses (377 plots).

The categorical data, i.e. country, climate zone, soil type and tree species are not included in this table

The species number per plot ranged from 1-127, with most data ranging between 10 and 80 species, whereas the Simpson index mostly ranged from 0.2-0.9. Tree age ranged mostly from 30 to 130 years with a fairly normal distribution. Since tree ages are given in 20 year, the mean age in each interval was taken. Stand height ranged mostly from 10 to 30 m., soil cover from 50-85% and altitude from 25 –1100 m. Altitude, precipitation and temperature are correlated variables. Altitude ranged from 25 m up to more than 1000 or 1800 m above sea level. The ranges in temperature were smaller than for precipitation and for most other variables because in most cases interpolated 30 year averaged data were used. Looking at the deposition values, the range between the 5 and 95 percentile is comparable for most ions. In general the 95 percentile is approximately 10-20 times as large as the 5 percentile. The element contents in the humus layer and mineral layer have a strongly skewed distribution, with the exception of C and N in the humus layer. The pH varies mostly between 3 and 5 in both the organic layer and mineral layer and mineral layer covers the whole range from 2-100%.

Part of the investigated variables are correlated. This information is relevant, since in deriving a robust model with relatively few environmental factors, it is crucial to delete those predictors that are strongly correlated with each other. Therefore the correlation coefficients between some of the predictors are presented here. As all predictors (except pH) were entered in the statistical analyses after taking the logarithm, these correlation coefficient have also been calculated after logarithmisation. The correlation between deposition variables is given in Table 3.10.

 Table 3.10
 Correlation matrix of the logarithmised bulk deposition variables. Correlation coefficients that are larger than 0.7 in absolute value are given in **bold**.

	Κ	Ca	Cl	Mg	Na	$NH_4$	$NO_3$
Ca	0.73	1.00					
Cl	0.49	0.52	1.00				
Mg	0.62	0.69	0.73	1.00			
Na	0.39	0.39	0.93	0.71	1.00		
$NH_4$	0.58	0.69	0.56	0.46	0.42	1.00	
NO <sub>3</sub>	0.45	0.61	0.59	0.47	0.50	0.83	1.00
$SO_4$	0.64	0.77	0.71	0.62	0.55	0.84	0.85

 Table 3.11
 Correlation matrix of the logarithmised predictors, excluding correlations between the deposition variables. Correlation coefficients that are larger than 0.4 in absolute value are given in **bold**. Predictors for which all correlation coefficients are smaller than 0.4 in absolute value have been omitted

	temp	Prec	Korg	Ca <sub>org</sub>	Mg <sub>org</sub>	pH <sub>org</sub>	P/C <sub>org</sub>	Bsat <sub>min</sub>	pH <sub>min</sub>	CEC <sub>min</sub>
Prec	0.13	1.00	-	-	-	-	-			
Korg	0.11	0.39	1.00							
Ca <sub>org</sub>	0.09	0.20	0.45	1.00						
Mg <sub>org</sub>	0.09	0.45	0.63	0.66	1.00					
pH <sub>org</sub>	0.28	0.19	0.52	0.72	0.69	1.00				
P/C <sub>org</sub>	-0.04	0.27	0.23	0.16	0.41	0.20	1.00			
Bsatmin	0.06	-0.13	0.26	0.53	0.32	0.62	0.00	1.00		
pH <sub>min</sub>	0.00	-0.05	0.27	0.37	0.36	0.59	0.09	0.58	1.00	
CEC <sub>min</sub>	0.06	0.51	0.43	0.41	0.58	0.36	0.22	0.11	0.08	1.00
K <sub>dep</sub>	0.44	-0.05	-0.09	-0.07	-0.09	0.04	-0.19	0.04	-0.02	-0.03
Ca <sub>dep</sub>	0.59	0.06	0.05	0.04	-0.05	0.11	-0.15	0.09	-0.01	0.04
Cl <sub>dep</sub>	0.56	0.41	-0.01	0.01	0.10	0.12	-0.02	-0.11	-0.10	0.19
Mg <sub>dep</sub>	0.43	0.25	0.01	0.10	0.16	0.14	-0.06	-0.01	0.01	0.06
Na <sub>dep</sub>	0.49	0.52	0.04	0.07	0.22	0.16	0.05	-0.14	-0.07	0.24
NH <sub>4,dep</sub>	0.71	0.16	-0.06	-0.12	-0.13	-0.04	-0.08	-0.13	-0.17	-0.02
NO <sub>3,dep</sub>	0.63	0.33	-0.03	-0.07	-0.06	-0.03	0.03	-0.20	-0.20	0.09
SO <sub>4,dep</sub>	0.64	0.19	-0.05	-0.11	-0.14	-0.04	-0.12	-0.11	-0.20	0.01

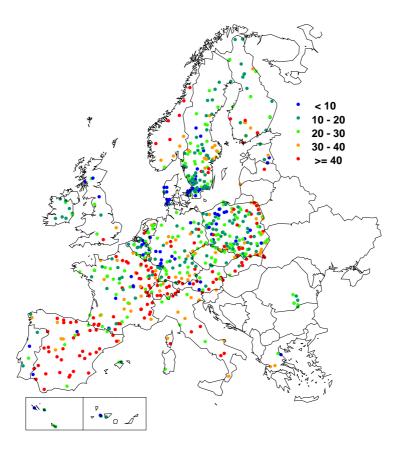
The results show that there is a very strong correlation between Na and Cl deposition, whereas the correlations between SO<sub>4</sub> and NO<sub>3</sub>, NH<sub>4</sub> and NO<sub>3</sub>, Ca and SO<sub>4</sub>, Cl and SO<sub>4</sub>, Mg and Na, Mg and Cl and Ca and K are also considerable (Table 3.10). These correlations indicate the impact of:

- Seasalt inputs (specifically the correlations between Na, Cl, Mg and to a lesser extent SO<sub>4</sub>);
- Atmospheric emissions, transport and chemistry determining NH<sub>4</sub>, NO<sub>3</sub> and SO<sub>4</sub> deposition;
- Soil dust (especially in dry climates, cf. the correlation between deposition and temperature in Table 3.11) probably causes the correlation between Ca and K.

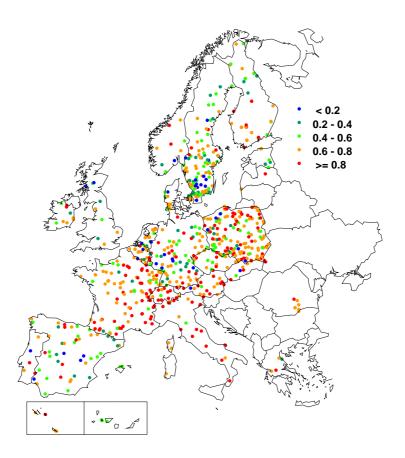
The most important correlations between nutrient contents in humus layer and mineral soil, including the variables indicating soil acidity are given in Table 3.11. The correlations between C, N and P in the humus layer have been reduced by using C/N and C/P ratios. Other expected correlations include those between pH in both layers, pH and base saturation in mineral layer and pH and total base cation concentration in humus layer. The correlation between soil variables (pH, base saturation, C/N ratio) and bulk deposition is, however, small.

#### 3.3.2 Geographical variation of species numbers, Simpson index and Ellenberg values

Maps of the number of species and the Simpson index for the 669 plots with >0 vascular species, using the original data are given in Figure 3.6 and 3.7, respectively.



*Figure 3.6* Species numbers for vascular plants at the 674 plots for which ground vegetation data were available up to 1999.



*Figure 3.7 Abundance weighted species diversity according to the Simpson index at the 674 plots for which ground vegetation data were available up to 1999.* 

The results show a North-South gradient with respect to species numbers with higher species numbers in the Mediterranean areas compared to the boreal forests, except for some plots in Norway. In Poland and France, the number of species increases from West to East. The Simpson index can vary strongly within countries and that there are no clear gradients over countries. As expected, the highest Simpson indices (high diversity) are associated with plots containing a high number of species (compare Figure 3.2) and very low Simpson indices (low diversity) with plots containing only a few species.

The North-South gradient in species richness is a well-known phenomenon (Lindeijer et al., 1998, Mucina, 1991), but in this case the steepness of the gradient may have been reduced by the omission of cryptogamic species, which make an important part of the vegetation of the North.

Figure 3.8 shows the Ellenberg indicator values for temperature and acidity for the 674 plots with ground vegetation data, respectively. The Ellenberg indicators reflect the well-known gradients in temperature and soil acidity over Europe. This is considered as an indication that the large-scale gradients in climate and soil over Europe are well represented by the plots, and that the Ellenberg indicator values to a certain degree reflect the response of the vegetation to these gradients.

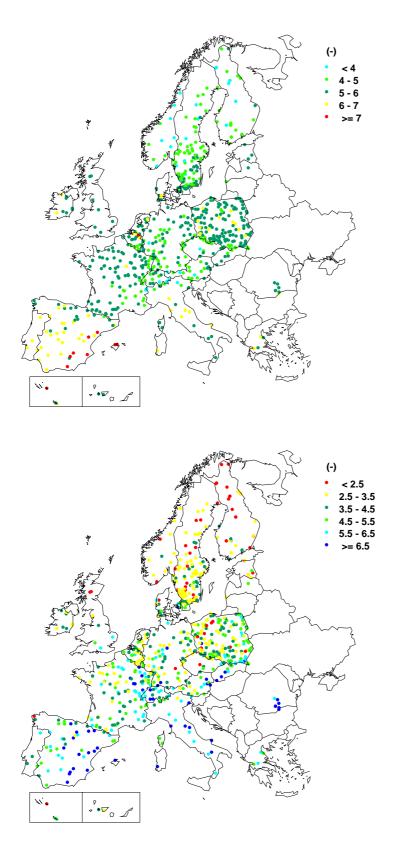


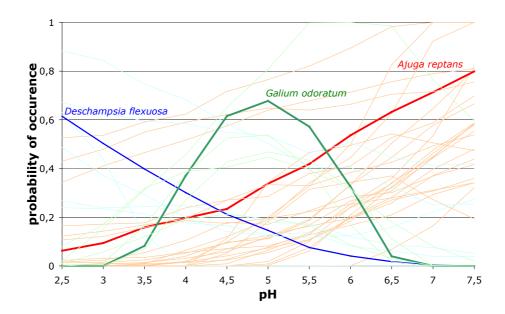
Figure 3.8 Ellenberg indicator values for temperature (Top) and acidity (Bottom) at the 674 plots for which ground vegetation data were available up to 1999.

# **3.3.3 Relationships between the occurrence probability of individual species and environmental factors**

## Response curves per species in dependence of soil factors and bulk deposition

With the valid regression formulae, a Monte-Carlo simulation has been executed to calculate the occurrence probability of 247 species by drawing 10,000 value combinations for all predictor variables. The predictor values were drawn independently from uniform distributions over the respective ranges encountered in the predictor dataset underlying the regression. The average occurrence probability of the different species was plotted against predictor values that were sub-divided into slices. Every value slice of a particular predictor variable is associated with the entire variation ranges in the other predictor variables.

In this way the response curves in Figures 3.9 to 3.13 are generated. The graphs are limited to the predictor variables that may be related to air pollution. In Figure 3.9 - 3.13, we only plotted those species in which the change in the maximum and minimum occurrence probability induced by the factor in consideration was more than 20%.



<u>Species that prevail in acid habitats (low pH) are</u>: **Deschampsia flexuosa**, Calluna vulgaris, Calamagrostis villosa, Vaccinium myrtillus, Vaccinium vitis-idaea, Picea abies, Sorbus aucuparia.

Species that prevail in intermediate habitatss are: Galium odoratum, Melica uniflora, Anemone nemorosa, Veronica officinalis, Hedera helix, Carex sylvatica.

<u>Species that prevail in alkaline habitats (high pH) are</u>: Ajuga reptans, Viola alba, Melittis melissophyllum, Dactylis glomerata, Sorbus domestica, Cardamine bulbifera, Silene italica, Digitalis lutea, Festuca heterophylla Daphne laureola, Cruciata glabra, Ruscus aculeatus, Carex flacca, Stachys officinalis, Rubus caesius, Poa nemoralis, Carpinus betulus, Mercurialis perennis, Solidago virgaurea, Rosa arvensis, Luzula forsteri, Rubus idaeus, Prunus spinosa, Rubus ulmifolius, Arum maculatum.

Figure 3.9 Response curves for species demonstrating a considerable response to the pH in the organic soil layer. The lines represent all species with a significant response, their names are enumerated below the graph. Three species that may be considered as examples of the three possible response types are represented as bold lines with their names added.

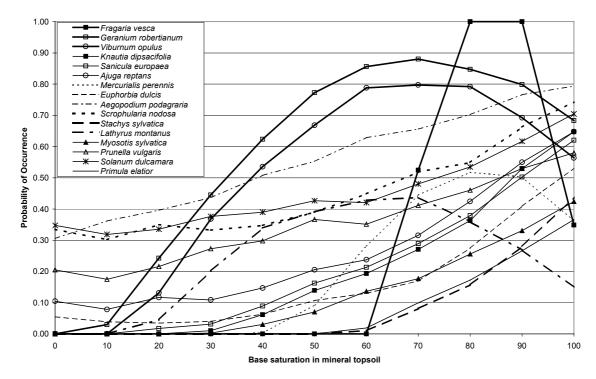


Figure 3.10 Response curves for species demonstrating a considerable response to base saturation (%).

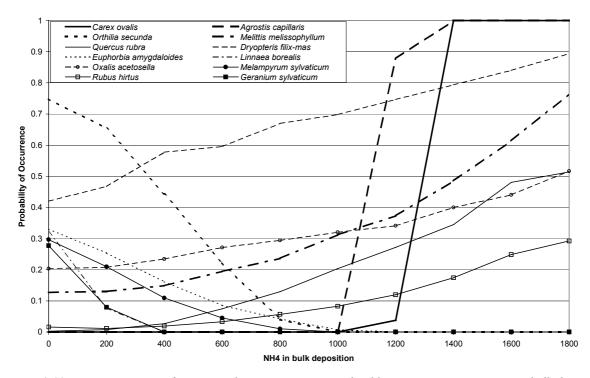


Figure 3.11 Response curves for species demonstrating a considerable response to ammonium in bulk deposition  $(mol_c.ha^{-1}.yr^{-1})$ .

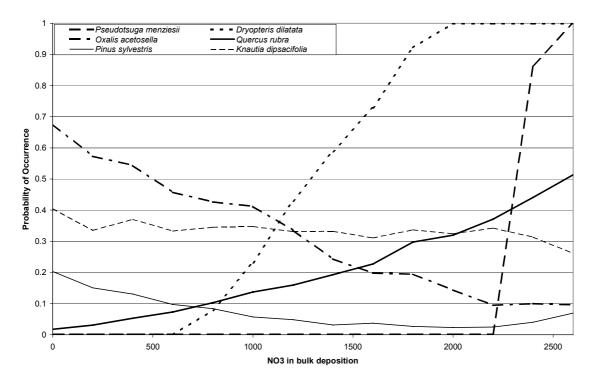


Figure 3.12 Response curves for species demonstrating a considerable response to nitrate in bulk deposition  $(mol_c.ha^{-l}.yr^{-l})$ .

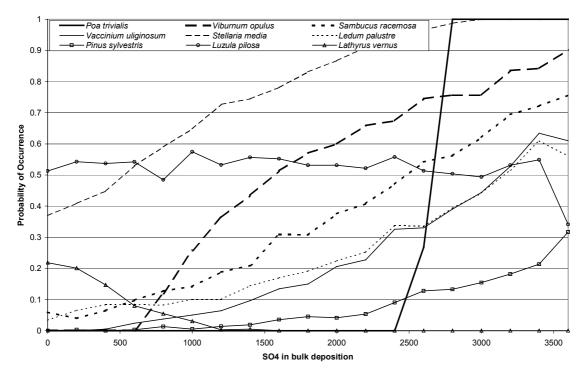


Figure 3.13 Response curves for species demonstrating a considerable response to the sulphate in bulk deposition  $(mol_c.ha^{-1}.yr^{-1})$ .

Out of the 332 considered species, this was the case for 36 species in the case of pH (Figure 3.9), 16 in case of base saturation (Figure 3.10), 12, 6 and 9 for  $NH_4$ ,  $NO_3$  and  $SO_4$  in bulk deposition, respectively. (Figure 3.11 - 3.13). In general, the derived response curves for pH agree quite well with the known ecology of the species (e.g., *Calluna vulgaris* decreasing toward higher pH values, *Ajuga reptans* increasing toward higher pH values), but they have no obvious ecological interpretation for the other predictors. It is surprising that only very few species show the unimodel response that is claimed in ecological textbooks; most species just increase with increasing pH.

## Derivation of indicator species

The regressions with the minimum set of predictors were also performed with standardised predictor values, i.e. after transformation according to  $Z' = (Z-MEAN(Z)] / \sqrt{(VAR[Z])}$ . As a result, the absolute magnitudes of the regression coefficients become comparable. Regression models were selected that are characterised by a more than average influence (absolute standardised regression coefficient value belonging to the top 10%) of the significant model predictors. An analysis of the most important predictor variables can identify indicator species in a qualitative manner, as is presented in Table 3.12. An explanation of the codes used in this table is given in Table 3.13. The indicator species are defined as species showing clear responses to variations in environmental factors. The regression coefficients marked black indicate a positive correlation between the occurrence of the species and the predictor value, while red regression coefficients indicate a negative correlation.

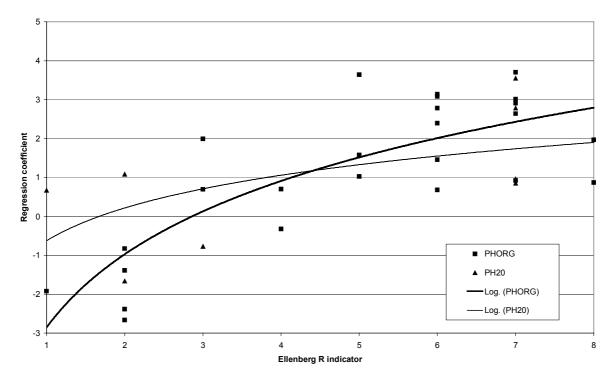


Figure 3.14 Ellenberg R-values against the regression coefficients for pH in organic and mineral topsoil. Dots: standardised regression coefficients; lines: fitted values of regression equations: regression coefficient = a + b \* Log(Ellenberg R) Regression equations: pH\_mineral = 1.2137 ln(R) -0.6249 and pH\_org = 2.7154 ln(R) -2.8546.

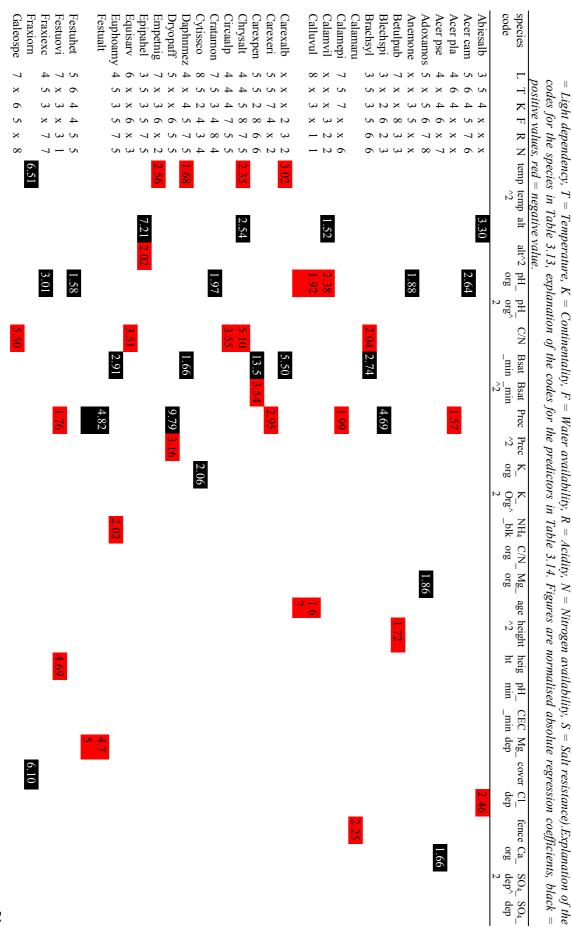
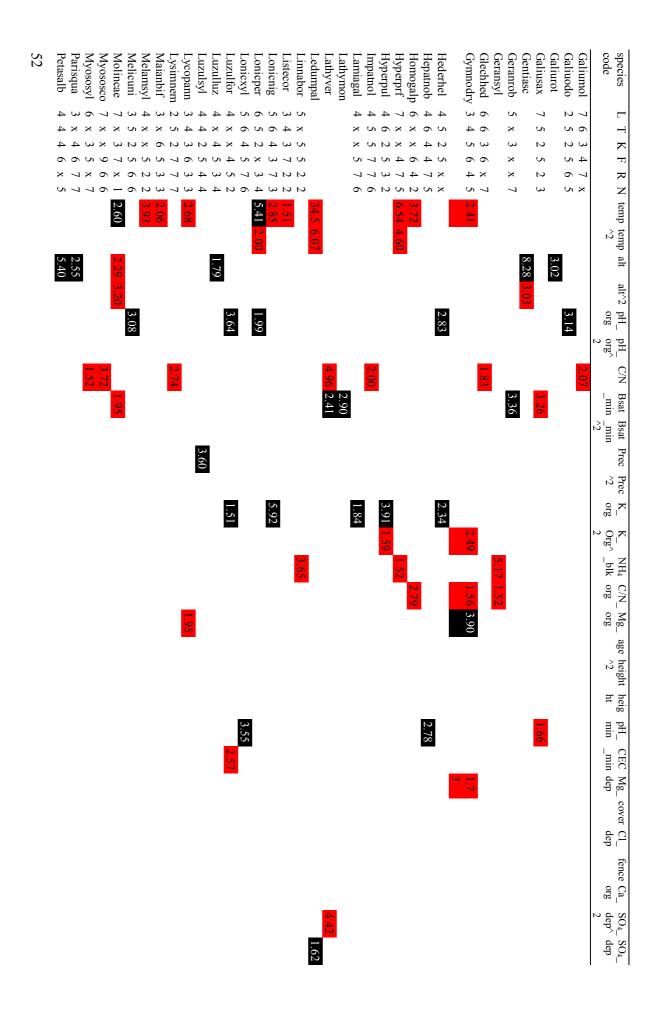
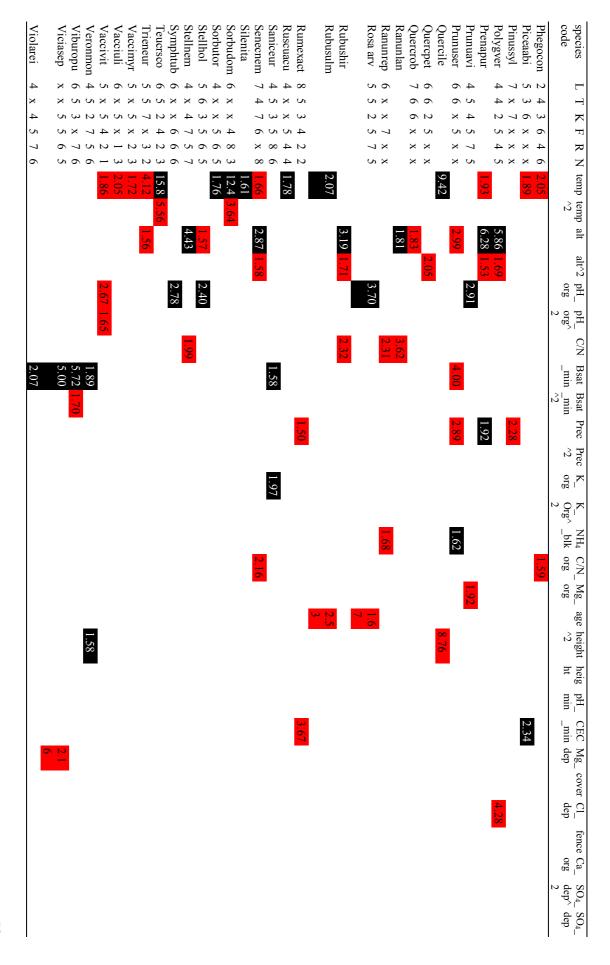


Table 3.12 Indicator species. Positive and negative response to a particular variable is indicated in black and red, respectively. The Ellenberg indicator values are also given (L





1	Explanation of species eo		<i>iele 5:12 unu 1 igui es 5:1</i> 0	1	
code	name	code	name	code	name
Abiesalb	Abies alba	Gentiasc	Gentiana asclepiadea	Ranunlan	Ranunculus lanuginosus
Acer cam	Acer campestre	Geranrob	Geranium robertianum	Ranunrep	Ranunculus repens
Acer pla	Acer platanoides	Geransyl	Geranium sylvaticum	Rosa arv	Rosa arvensis
Acer pse	Acer pseudoplatanus	Glechhed	Glechoma hederacea	Rubusfru	Rubus fruticosus
Adoxamos	Adoxa moschatellina	Gymnodry	Gymnocarpium	Rubushir	Rubus hirtus
Agroscap	Agrostis capillaris		dryopteris	Rubusida	Rubus idaeus
Ajugarep	Ajuga reptans	Hederhel	Hedera helix	Rubussax	Rubus saxatilis
Anemonem	Anemone nemorosa	Hepatnob	Hepatica nobilis	Rubusulm	Rubus ulmifolius
Athyrfil	Athyrium filix-femina	Homogalp	Homogyne alpina	Rumexact	Rumex acetosella
Betulpen	Betula pendula	Hyperprf	Hypericum perfoliatum	Ruscuacu	Ruscus aculeatus
Betulpub	Betula pubescens	Hyperpul	Hypericum pulchrum	Samburac	Sambucus racemosa
Blechspi	Blechnum spicant	Ilex aqu	Ilex aquifolium	Saniceur	Sanicula europaea
Brachsyl	Brachypodium	Impatnol	Impatiens noli-tangere	Senecnem	Senecio nemorensis
	sylvaticum	Impatpar	Impatiens parviflora	Silenita	Silene italica
Brizamax	Briza maxima	Junipcom	Juniperus communis	Solidvir	Solidago virgaurea
Calamaru	Calamagrostis	Lamiagal	Lamiastrum	Sorbuauc	Sorbus aucuparia
	arundinacea	e	galeobdolon	Sorbudom	Sorbus domestica
Calamepi	Calamagrostis epigejos	Lathymon	Lathyrus montanus	Sorbutor	Sorbus torminalis
Calamvil	Calamagrostis villosa	Lathyver	Lathyrus vernus	Stellhol	Stellaria holostea
Calluvul	Calluna vulgaris	Lavensto	Lavandula stoechas	Stellnem	Stellaria nemorum
Carexalb	Carex alba	Ledumpal	Ledum palustre	Symphtub	Symphytum tuberosum
Carexeri	Carex ericetorum	Ligusvul	Ligustrum vulgare	Teucrsco	Teucrium scorodonia
Carexpen	Carex pendula	Linnabor	Linnaea borealis	Trieneur	Trientalis europaea
Carexpil	Carex pilulifera	Listecor	Listera cordata	Tubergut	Tuberaria guttata
Carexsyl	Carex sylvatica	Lonicnig	Lonicera nigra	Urticdio	Urtica dioica
Carpibet	Carpinus betulus	Lonicper	Lonicera periclymenum	Vaccimyr	Vaccinium myrtillus
Chrysalt	Chrysosplenium	Lonicxyl	Lonicera xylosteum	Vacciuli	Vaccinium uliginosum
Chrysan		Luzulfor	-	Vaccivit	Vaccinium uitginosum Vaccinium vitis-idaea
Cimeral	alternifolium Cinanan alminan		Luzula forsteri		
Circaalp	Circaea alpina	Luzulluz	Luzula luzuloides	Veronmon	Veronica montana
Convamaj	Convallaria majalis	Luzulpil	Luzula pilosa	Veronoff	Veronica officinalis
Corylave	Corylus avellana	Luzulsyl	Luzula sylvatica	Viburopu	Viburnum opulus
Cratamon	Crataegus monogyna	Lycopann	Lycopodium annotinum	Viciasep	Vicia sepium
Cytissco	Cytisus scoparius	Lysimnem	Lysimachia nemorum	Violarei	Viola reichenbachiana
Daphnmez	Daphne mezereum	Maianbif	Maianthemum bifolium	Violariv	Viola riviniana
Deschces	Deschampsia cespitosa	Melampra	Melampyrum pratense		
Deschfle	Deschampsia flexuosa	Melamsyl	Melampyrum sylvaticum		
Dryopaff	Dryopteris affinis	Melicuni	Melica uniflora		
Dryopcar	Dryopteris carthusiana	Miliueff	Milium effusum		
Dryopdil	Dryopteris dilatata	Moehrtri	Moehringia trinervia		
Dryopfil	Dryopteris filix-mas	Molincae	Molinia caerulea		
Empetnig	Empetrum nigrum	Mycelmur	Mycelis muralis		
Epiloang	Epilobium	Myososco	Myosotis scorpioides		
	angustifolium	Myososyl	Myosotis sylvatica		
Epipahel	Epipactis helleborine	Oxaliace	Oxalis acetosella		
Equisarv	Equisetum arvense	Parisqua	Paris quadrifolia		
Ericacin	Erica cinerea	Petasalb	Petasites albus		
Euphoamy	Euphorbia	Phegocon	Phegopteris connectilis		
	amygdaloides	Piceaabi	Picea abies		
Fagussyl	Fagus sylvatica	Pinussyl	Pinus sylvestris		
Festualt	Festuca altissima	Poa nem	Poa nemoralis		
Festuhet	Festuca heterophylla	Polygver	Polygonatum		
Festuovi	Festuca ovina		verticillatum		
Fragaves	Fragaria vesca	Prenapur	Prenanthes purpurea		
Frangaln	Frangula alnus	Prunuavi	Prunus avium		
Fraxiexc	Fraxinus excelsior	Prunuser	Prunus serotina		
Fraxiorn	Fraxinus ornus	Pteriaqu	Pteridium aquilinum		
Galeospe	Galeopsis speciosa	Querccer	Quercus cerris		
Galiumol	Galium mollugo	Quercile	Quercus ilex		
Galiuodo	Galium odoratum	Quercpet	Quercus petraea		
Galiurot	Galium rotundifolium	Quercrob	Quercus robur		
Galiusax	Galium saxatile	Quercrub	Quercus rubra		
5.4			~		

 Table 3.13
 Explanation of species codes used in Table 3.12 and Figures 3.16 - 3.19

The species requirements as identified by the regression coefficients in Table 3.12 have been compared with the Ellenberg indicator values. As is illustrated in Figure 3.14, the pH in both organic and mineral topsoil are the only predictors producing a consistent correspondence with the associated Ellenberg indicator (R). Low Ellenberg values, indicative for acid conditions, are associated with negative regression coefficients for pH, indicating that species with a low Ellenberg R (acidophytic species) are decreasing with increasing pH. Similarly, high Ellenberg R values are associated with positive regression coefficients, indicating an increase of neutrophytic species with increasing pH.

# 3.3.4 Relationships between the species composition and environmental factors

## Explained variance of the species composition by environmental factors

In CCA, various statistical models were tested to derive a model that explains a maximum amount of variance using a minimum number of predictor variables. In doing so, two criteria were used: the F-value (ratio between the extra variance explained by the model and residual variance), and the P-value (probability that the effect of a variable is due to coincidence). Three of these models are considered here: the 'full' model (that uses all available predictors), the 'significant' model (that uses the predictors for which F > 1.5 and P < c. 0.1) and the 'restricted' model (with only the predictors for which F > 2 and P < c. 0.01). As the P values were derived by bootstrapping, the values given are samples drawn from a population with a certain spread (which decreases as the number of bootstrap samples increases), and should therefore not be interpreted too strictly. All models shown in this section were derived using the 360 plots selected in Table 3.7 and the 396 occurring  $\geq 3$  times in these plots.

Table 3.14 compares the three models given above, and also compares the variance explained by the countries with the variance explained by the environmental predictors.

predictor set	percentage explained variance	number of predictors
all predictors <sup>1</sup> )	32%	64
only countries	13%	20
only environmental variables	24%	47
uniquely due to environmental variables	19%	
uniquely due to countries	7%	
undetermined	5%	
full model (countries as covariables)	19%	44 <sup>2</sup> )
significant model	14%	$24^{2}$ )
restricted model	10%	$12^{2}$

Table 3.14 Overview of total percentages explained variance for different models in CCA.

<sup>1</sup>) see Table 3.6 for a complete list of variables

<sup>2</sup>) plus 20 covariables to account for the effect of the countries

Out of a total of 32% variance explained by the complete model (using all available predictors), 7% is uniquely due to the effect of the countries. This may be considered as an indication that methodological differences cause a considerable bias in the data, even though country does include other aspects than methodological differences only (compare Klap et al., 2000, who found similar results when relating environmental factors to crown condition). To adjust for this bias, the countries have been used as 'co-variables' in all subsequent CCA analyses, i.e., their effect

was accounted for before calculating tables of explained variance and before drawing biplots. From Table 3.14 it may be seen that 19% of the total variance is uniquely due to the effect of the 'real' environmental variables (i.e., excluding the countries as predictors), so this is also the amount of variance accounted for by the full model after adjusting for the effect of country. In interpreting these figures it should be borne in mind that in CCA on ecological data, percentages explained variance between 10 and 20% are quite usual (Jongman et al., 1995).

Table 3.15 shows the result of a forward selection in CCA, and the variables that were in the 'significant' and the 'restricted' model. In the 'significant' model the correlation between the quantitative explanatory variables is  $r \le 0.5$  with one exception (pH\_min and pH\_org, r=0.59), which is judged acceptable.

	Variable <sup>1</sup> )	Р	F	% variance explained	% variance explained
				explained	(cumulative)
	pH_org	0.001	7.80	1.94%	1.94%
	spruce	0.001	4.37	1.11%	3.05%
	beech	0.001	4.50	1.11%	4.15%
e.	Mediterranean low	0.026	3.30	0.83%	4.98%
pot	Continental	0.022	3.09	0.74%	5.72%
1, 1	Atlantic south	0.018	2.96	0.74%	6.46%
stec	pH min	0.005	2.91	0.65%	7.11%
'restricted' model	N/C min	0.003	2.69	0.65%	7.75%
,re,	oak	0.022	2.33	0.55%	8.31%
del	K org	0.008	2.19	0.55%	8.86%
noc	Ca org	0.008	2.19	0.46%	9.32%
'significant' model	temp	0.016	2.10	0.55%	9.88%
car	mountain south	0.032	1.95	0.46%	10.34%
nifi	south	0.034	1.89	0.46%	10.80%
Sig	pine	0.038	1.89	0.37%	11.17%
	N/C org	0.031	1.76	0.46%	11.63%
	Cambisol	0.017	1.70	0.37%	12.00%
	Bsat min	0.019	1.71	0.37%	12.37%
	altitude	0.048	1.62	0.37%	12.74%
	P/C org	0.058	1.55	0.37%	13.11%
	Mediterranean high	0.077	1.50	0.37%	13.47%
	precipitation	0.065	1.40	0.37%	13.84%
	Na dep	0.044	1.57	0.28%	14.12%
	NO3 dep	0.076	1.40	0.37%	14.49%
	CEC min	0.106	1.32	0.28%	14.77%
	age	0.131	1.31	0.28%	15.04%
	Luvisol	0.166	1.25	0.37%	15.41%
	(further terms not gi	ven)			

Table 3.15 Forward selection of variables in CCA. P = probability of this, or a higher F-value under the null hypothesis as determined on the basis of 999 bootstrap samples; F = (regression sum of squares with this term - regression sum of squares without this term) / error mean square.

<sup>T</sup>) \_min = in mineral layer; \_org = in organic layer; \_dep = in bulk deposition; Bsat = base saturation; spruce, beech, pine = tree species; Mediterranean low, continental, Atlantic south, mountain south, south, Mediterranean high = climates; Cambisol, Luvisol = soil types

A summary of the significant model is given in Table 3.16. In the analysis presented until now, only bulk deposition has been used as the indicator for deposition because for this variable most observations were available, leading to a number of 360 plots. However, an extra analysis with a smaller number of plots (194 plots) has also been carried out to explore the effect of using total deposition or throughfall instead of bulk deposition. The main results of that analysis are also

given in Table 3.16. Results show that the explained variance is for a large part due to the actual situation of the soil, specifically soil acidity, climate and tree species. When using bulk deposition chemistry, only a small portion of the explained variance (0.7% out of the 14.5% explained variance of the significant model) is due deposition. In the evaluation with throughfall, the explained variance is better and comes to a total of 3.3% (out of the 20.7% explained variance). In this context, one has to realise that Na deposition (which plays a role when using bulk deposition chemistry) and K in throughfall are both of natural origin. In case of K it is due to recycling by soil uptake and foliar excretion by trees. The effect of Na may be partly artificial because its deposition is strongly correlated to the distance to the coast, and may therefore be just an indicator for a climatic effect that is not accounted for in our 'climate' variables. It are only NH<sub>4</sub> and NO<sub>3</sub> that are anthropogenic origin and these ions explain only 0.4 to 1.2% of the explained variation, depending upon the kind of analysis. Inversely, one has to be aware that direct effect acid deposition can partly be hidden in the 'actual soil acidity' which is most likely influenced by deposition in the past. The analysis with throughfall and total deposition is described in more detail in Annex 2.

 Table 3.16
 Summary of effect of variables in the 'significant' models using various selections of plots and deposition variables. Figures are percentages explained variance.

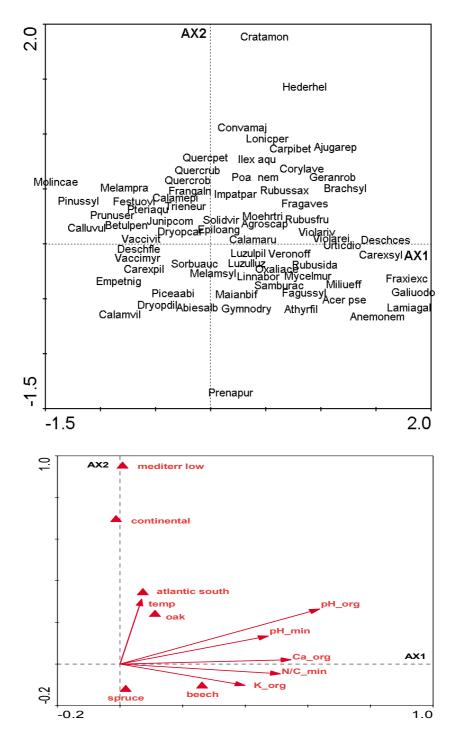
Variable group	360 plots, bulk	194 plots, bulk or 194 plots,		
	deposition	total deposition	throughfall	
Actual soil situation	5.8%	7.8%	7.6%	
Climate <sup>1</sup>	4.9%	6.1%	5.6%	
Tree species	3.1%	4.9%	4.1%	
Deposition: non-anthropogenic (K, Na)	0.3%	0.0%	2.1%	
Deposition: anthropogenic (NH <sub>4</sub> , NO <sub>3</sub> )	0.4%	0.0%	1.2%	
SUM	14.5%	18.7%	20.7%	

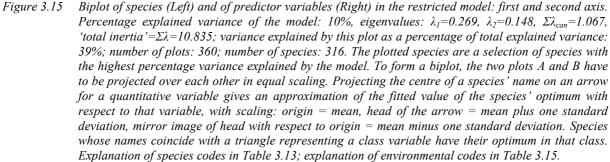
<sup>1</sup> Includes climate zone, altitude, temperature, precipitation

# Evaluation of results in biplots

Figure 3.15 is the biplot of first and second axis of the restricted model. When the ecology of the species is considered, a circular gradient of forest types can be seen in the species part of the biplot, which is depicted in Figure 3.16. Figure 3.17 is the biplot of the third en fourth axis of the restricted model. Figure 3.18 is a biplot that represents the effect of Na and NO<sub>3</sub> in bulk deposition. To facilitate the ecological interpretation of the biplots, the sample scores on the axes have been regressed on the average Ellenberg indicator values per plot. The results of these analyses are given in Tables 3.17 (for the restricted model) and 3.18 (for the effect of the deposition variables in the significant model).

By combining Figures 3.15, 3.16 and 3.17, and Table 3.15, a general picture can be formed of the principal directions of variation in the species data and their most probable causes. It should be kept in mind that the axes are ordered to decreasing importance, i.e. the most important direction of variation is represented along the first axis. This axis separates forests of rich soils, with species like *Corylus avellana, Fraxinus excelsior, Galium odoratum, Viola riviniana* agg., *Anemone nemorosa* from forest of poor soils with species like *Pinus sylvestris, Calluna vulgaris, Vaccinium* spp., *Trientalis europaea*.





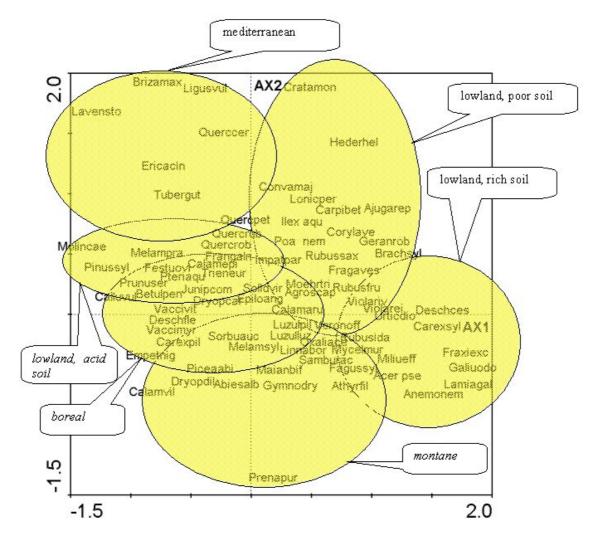


Figure 3.16 Indication of the climate and soil preference of the species in the arrangement of Figure 3.16, based on the species' ecology and Ellenberg indicator values. Some of the rarer Mediterranean species with a low percentage of explained variance that do not appear in Figure 3.16 have been added.

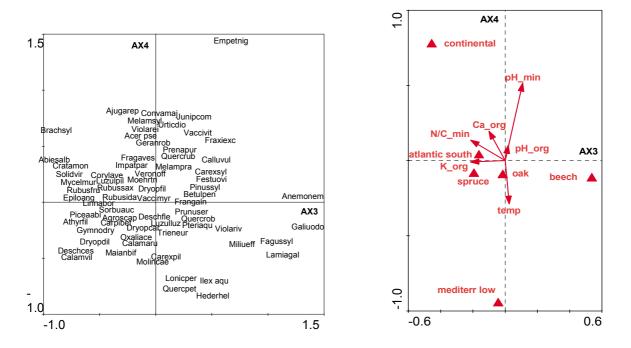


Figure 3.17 Biplot of species (A) and of predictor variables (B) in the restricted model: third and fourth axis. eigenvalues:  $\lambda_3 = 0.123$ ,  $\lambda_4 = 0.097$ ; variance explained by this plot as a percentage of total explained variance: 21%. Further explanation as in Figure 3.16.

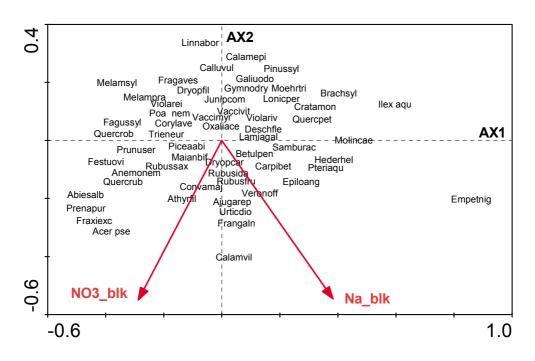


Figure 3.18 Biplot of species against predictors  $NO_3$  and Na in bulk deposition, after adjustment for the effect of country and all environmental variables of the significant model except  $NO_3$  and Na. Percentage explained variance of the model: 0.8%. Eigenvalues:  $\lambda_3 = 0.054$ ,  $\lambda_4 = 0.028$ ; 'total inertia' =  $\Sigma\lambda$  = 10.835; variance explained by this plot as a percentage of total explained variance: 100%. Further explanation as in Figure 3.16.

The principal ecological determinants of 'rich soil forests' are the predictors with a positive score on the first axis, i.e. pH (both in mineral and organic layer), basic cations (Ca and K), and N/C ratio. This interpretation is confirmed by the analysis of Ellenberg values (Table 3.17), where acidity and nutrient availability are positively correlated with the sample score on the first axis. The indicator for light availability is strongly negatively correlated with the first axis. Although no light measurements are available, it is probable that this relation reflects the light climate, which tends to be darker in forests on rich soil.

The second axis separates the mountain forests from those in the lowland. At the lower end of this axis are typical mountain species like *Abies alba, Praenanthes purpurea, Calamagrostis villosa,* while at the higher end are lowland (Atlantic or Mediterranean) species like *Ilex aquifolium, Hedera helix, Erica cinerea* and *Lavandula stoechas* (the Mediterranean species are only in Figure 3.17). This interpretation is confirmed by the environmental scores in the biplot: temperature, and the climate classes 'Mediterranean low' and 'Atlantic south' have high scores on the second axis. Surprisingly, the Ellenberg temperature indicator has no positive relation with this axis. According to their Ellenberg values, the mountain species at the lower end of this axis have a preference for shade and relatively wet, acid and nutrient-rich conditions.

The third axis is mainly related to tree species. This axis separates beech forests (with species like *Galium odoratum, Anemone nemorosa, Lamium galeobdolon*) at the positive end, from spruce forests (with species like *Gymnocarpium dryopteris* and other ferns, *Solidago virgaurea, Linnea borealis*) at the negative end. Oak forests take an intermediate position. Pine is left out because of its too low contribution to explained variance (although its position can be guessed from the position of the 'pine-forest-species'). Ellenberg's temperature indicator is strongly positively correlated with this axis, while the light and acidity indicators are negatively correlated. This is understandable on the basis of the soil pH, temperature preference and light climate of oak, spruce and beech forests, which have a gradient in the direction less acid, warmer, darker, in the order pine, oak, beech.

Table 3.17	Regression coefficients of the regression of sample scores of the restricted model (i.e., the model depicted
	in Figures 3.16, 3.17 and 3.18) on Ellenberg's indicator values. The values can be interpreted as the
	expected distance along a given axis between species whose values for a given indicator differ by one
	unit. Significance classes: *** = $P \le 0.001$ , **= $0.001 < P \le 0.01$ , *= $0.01 < P \le 0.05$ , <sup>ns</sup> = $P > 0.05$ . %expl
	(indicator) is the percentage variance explained by the regression of sample scores on indicator values,
	% expl (species) is the percentage variance explained by each axis in CCA (i.e., $\lambda_n / \Sigma \lambda$ ), % expl (plot) is
	the percentage variance explained by each axis in CCA relative to the total amount of explained variance
	$(i.e., \lambda_n / \Sigma \lambda_{can}).$

$(1.c., \lambda_n / \Delta \lambda_{can})$ .							
indicator	AX1	AX2	AX3	AX4			
light	-0.39***	1.04***	-0.76***	0.08 <sup>ns</sup>			
temperature	-0.22*	0.15 <sup>ns</sup>	0.81***	-0.04 <sup>ns</sup>			
continentality	0.33**	$-0.38^{ns}$	0.10 <sup>ns</sup>	0.96***			
humidity	$0.09^{ns}$	-0.53**	$-0.03^{ns}$	$0.27^{ns}$			
acidity	0.54***	1.03***	-0.34**	$0.17^{ns}$			
nutrient availability	0.23**	-0.84***	-0.11 <sup>ns</sup>	0.04 <sup>ns</sup>			
%expl (indicator)	51.5%	37.8%	9.9%	4.7%			
%expl (species)	2.5%	1.4%	1.1%	0.9%			
%expl (plot)	25.2%	13.9%	11.5%	9.1%			
eigenvalue	0.269	0.148	0.123	0.097			

The fourth axis is related to climate. Here the continental species like *Empetrum nigrum, Ledum palustre, Asarum europaeum* (the latter two not plotted) are at the positive end, while Atlantic species like *Ilex aquifolium, Hedera helix, Erica arborea, Ruscus aculeatus* (the latter two not

plotted) are at the negative end. Continentality is the only indicator that is significantly related to this axis. Of the environmental predictors, temperature and the climate zone 'Mediterranean lower' have a strongly negative score on this axis, while the climate zone 'continental' has a strongly positive score.

The arrangement of the species in Figure 3.19 is less easy to interpret. In the lower half of the plot (i.e., with a low score on the second axis) are a number of species that preferentially occur in wet forests, like *Fraxinus excelsior, Urtica dioica, Anemone nemorosa.* These are also species that prefer nutrient rich circumstances occurring in e.g. river shorelands. In the upper half of the plot are species preferring drier conditions, both nutrient-poor (like *Vaccinium vitis-idea, Calluna vulgaris)* and nutrient-rich (like *Galium odoratum, Viola reichenbachiana)*. This interpretation is confirmed by the Ellenberg indicator values, where the humidity indicator is strongly negatively related to the second axis. It is difficult to discover a consistent pattern in the species' ecology with respect to the first axis. However, the relation with the indicator values (Table 3.18) reveals an increase of species preferring nutrient-rich sites, and of continental species, towards the lower end of the first axis. This is exactly the pattern that would be expected on the basis of the predictor scores on this axis (the positive score of Na in bulk deposition indicates more oceanic conditions at the upper end of this axis, and thus more continental conditions at the lower end). Therefore the effect of the deposition variables as found in the multivariate analysis probably represents a real effect, although the statistical significance is only weak.

Table 3.18 Regression coefficients of the regression of sample scores of a model containing terms for  $NO_3$  and Na in bulk deposition, after adjustment for the effect of country and all environmental variables of the significant model except  $NO_3$  and Na (i.e., the model depicted in Figure 3.19). Further explanation as in Table 3.17

Table 3.1	Table 3.17.								
indicator	AX1	AX2							
light	1.14***	0.39 <sup>ns</sup>							
temperature	-0.33 <sup>ns</sup>	$-0.10^{ns}$							
continentality	-1.98***	-0.45 <sup>ns</sup>							
humidity	-0.41 <sup>ns</sup>	-1.19***							
acidity	0.48 <sup>ns</sup>	-0.52*							
nutrient availability	-1.01***	0.02 <sup>ns</sup>							
%expl (indicator)	19.9%	10.1%							
%expl (species)	0.5%	0.3%							
%expl (plot)	65.9%	34.1%							
eigenvalue	0.054	0.028							

## Analysis of biodiversity measures

Table 3.19 shows various models resulting from the regression of the logarithmised species numbers on the predictors included in the significant model and the restricted model as given in Table 3.15, and the deposition variables. The total percentages of explained variance are approximately equal for all models (c. 40%) except the restricted model without deposition terms which has an appreciably lower amount of explained variance (c. 30%). Table 3.20 shows models derived in the same fashion with the Simpson index as the dependant variable. Here the percentages of explained variance are lower but again only slightly different between the models (c. 17%) except for the restricted model without deposition (c. 14%). Both the species numbers and the Simpson index can be quite well explained by models without any deposition terms; the Simpson index is even best explained by a simple model that only contains terms for pH, tree species, base saturation and altitude. If deposition terms are included in a model, these are nearly always the ions that originate from soil or seawater.

Table 3.19 Regression of log(species number) on the predictors of the significant and the restricted model in Table 3.15. Explanation of the predictor terms in Table 3.15. All models are minimal models derived by backward selection of the terms from the models in Table 3.15 until only terms remain that significantly contribute to the fit of the model (criterion:  $|t_{regression \ coefficient}| \leq 0.05$ ). +depo: model resulting from the addition of all deposition terms (Table 3.6) to the minimal model in the previous column, followed by backward selection until  $|t| \leq 0.05$  for all terms. The sign of the figures in column 'signif' is the sign of the regression coefficient, the absolute value refers to its significance:  $3 = t \leq 0.001, 2 = 0.001 < t \leq 0.01, 1 = 0.01 < t \leq 0.05$ . The figures in the column '%expl' is the percentage explained variance that is exclusively due to each term (i.e., the drop in percentage explained variance on removing this term from the model). The row 'undetermined' gives the percentage explained variance that is not exclusively attributable to any single term, 'total' is the total percentage explained variance. - = term with no significant effect, not in the model. Number of observations: 360.

	sign. m	odel	sign.mo	d.+depo	restr.mo	odel	restr.mod.+depo	
Variable	signif	%expl	signif	%expl	signif	%expl	signif	%expl
pH_org	2	1.5%	2	1.7%	3	7.0%	3	6.9%
beech	-3	3.9%	-3	2.3%	3	3.0%	-3	1.8%
Atlantic south	2	1.2%	2	1.2%	-	-	-	-
K_org	2	1.1%	3	2.0%	3	2.6%	3	2.6%
mountain south	3	2.0%	1	0.7%	-	-	-	-
south	3	4.0%	-	-	-	-	-	-
Bsat_min	3	5.7%	3	3.1%	-	-	-	-
altitude	2	1.3%	-	-	-	-	-	-
NO <sub>3</sub> _dep	-	-	-	-	-	-	-2	1.5%
K_dep	-	-	2	1.2%	-	-	1	0.7%
Ca_dep	-	-	3	3.9%	-	-	3	4.9%
Cl dep	-	-	-1	0.7%	-	-	-	-
Mg_dep	-	-	-3	2.3%	-	-	-3	5.3%
Undetermined		18.6%		23.8%		10.9%		14.2%
Total		39.3%		42.8%		29.7%		39.0%

 Table 3.20
 Regression of the Simpson index on the predictors of the significant and the restricted model in Table

 3.15. Further explanation as in Table 3.19

	sign.model		sign.mod.+depo		restr.model		restr.mod.+depo	
variable	signif	%expl	signif	%expl	signif	%expl	signif	%expl
pH_org	2	1.6%	-	-	3	5.1%	3	5.4%
pH_min	-1	0.8%	-	-	-	-	-	-
oak	2	1.5%	3	3.0%	1	1.2%	2	1.4%
Bsat_min	1	1.2%	3	8.4%	-	-	-	-
altitude	2	1.6%	3	3.2%	-	-	-	-
Ca_dep	-	-	-	-	-	-	3	2.7%
Mg_dep	-	-	-	-	-	-	-1	1.1%
NH <sub>4</sub> _dep	-	-	3	3.1%	-	-	-	-
Undetermined		8.7%		-0.4%		4.4%		5.3%
Total		17.5%		17.2%		14.8%		17.3%

The anthropogenic ions  $NO_3$  and  $NH_4$  have a significant effect in only one model each: for  $NO_3$  this is a negative effect on the species number, and for  $NH_4$  a positive effect on the Simpson index. None of the models shows a significant effect of  $SO_4$ .

The pH of the organic layer seems to be the most important factor determining the floristic diversity. This term has a positive effect in nearly all models. Next important is probably tree species, where beech has a negative effect on species number in all models except one, and oak has a positive effect on the Simpson index in all models. Two indicators for nutrient availability, base saturation and potassium in the organic layer, also appear in many of the models, with a positive effect on both species number and Simpson index. Southern climates and high altitude have a positive effect on species number or Simpson index in some of the models. Of the non-

anthropogenic ions in deposition, the seawater-related ones (Mg and Cl) have a negative effect on both species number and Simpson index, while the soil-related ones (Ca and K) have a positive effect.

# **3.4 Discussion and conclusions**

# 3.4.1 Univariate regression analyses on individual species

Though highly significant, median explained deviance of the models constructed is only about 30%. On a continental scale, the occurrence of plant species is obviously governed by additional factors. For a rather large number of species, the regression analysis produces significant information on species environment interactions with respect to a variety of factors. The lack of correspondence between the Ellenberg indicator values and the regression output needs to be studied in more detail. In the coming year, the data and the regression models need to be further elaborated. The results of the multivariate approach and the regression analysis must be compared, which may lead to further model refinement in both methodologies.

Future studies should address the following alternatives:

- 1. In this study the data on the species composition of the ground vegetation at "Intensive Monitoring" plots(ICP-F) have not yet been merged with those at "Integrated Monitoring" plots (ICP-IM). It is also questionable whether this is the most appropriate way. It is intended to use the ICP-IM data for validation of the models that are constructed with the Intensive monitoring data.
- 2. The stepwise procedure used in the preliminary analysis presented in the present paper is extremely in favour of "small" regression models (low number of predictors). It is to be tested if a less stringent approach is not leading to more representative models.

# 3.4.2 Multivariate correspondence analysis on the species composition

# Relation between species composition and stand and site factors

The multivariate analysis of the present data indicates that forest vegetation over Europe is mainly determined by the actual soil acidity status, climate in terms of precipitation and temperature, and tree species. Although the effect of bulk deposition is statistically significant, its contribution to the fit of a model is very low. The statistical approach may, however, have hidden the real deposition effects, as discussed below in the section on deposition effects. The biplots in Figures 3.16 and 3.17 show a gradient in species composition that generally agrees with the species' ecology and distribution pattern as shown in Flora Europaea (Tutin et al., 1964-1980) and local floras (e.g., Fournier, 1990; Oberdorfer, 1977, 1978, 1983; Lid, 1987). In multivariate statistics the predictors associated with the subsequent axes indicate their order of decreasing importance; in this case these predictors can be summarised as soil chemistry for the first axis, climate (mainly the altitudinal gradient) for the second axis, tree species (and climate, mainly the north - south gradient) for the third axis, and again climate (mainly the east - west gradient) for the fourth axis. This confirms the conclusion that can be drawn from Table 3.15, namely that soil, climate and tree species as the most important factors that determine the composition of ground vegetation, whereby their importance decreases in the order given. In general, these conclusions strongly support the ideas on species - environment relationship as found phytosociological literature (Oberdorfer, 1977, 1978, 1983; Braun-Blanquet, 1964). 64

# Impact of country

The high percentage variance that is explained by the countries is an indication that a considerable amount of 'noise' is introduced by methodological differences between the countries. If there had been more uniformity in these methods the effect of predictors that vary on a regional scale such as climate, soil type or deposition, would have been more pronounced. In that case the inclusion of the country as covariables would become unnecessary. As stated in Section 3.2.2, "country" does, however, not only include the impact of different data assessment methods but also different ecological circumstances that we could not include in the analysis. This includes differences in history (forest-degradation, plantation activities, historical deposition, legislation etc.), but also the differences in present deposition in climate variables between the countries. The applied approach implies that the within-country variation of the considered environmental factors is included and combined for all the countries considered, i.e., the same linear relationship between vegetation and deposition is assumed for each country, with a bias that is different per country. Limitation of this approach has been discussed in Klap et al. (2000), who used a comparable approach in relating tree crown condition to environmental variables. The inclusion of country thus only gives a rough impression of the possible impact of methodological differences between countries. In the future, the impact of real methodological differences should be assessed, based on e.g. field comparison by assessment teams. Preferably, registration of forest management at each site should be mandatory and sampling methods should be made uniform.

# Impacts of atmospheric deposition

The 'significant' model contains two bulk deposition terms, one of which is of non-anthropogenic origin (Na). As the deposition variables are strongly correlated, their terms should be seen as indicators for groups of variables rather than as real causal agents. In the correlation matrix, three of such groups can be identified: seawater-derived (Na, Cl, Mg), soil-derived (Ca, K) and anthropogenically-derived (SO<sub>4</sub>, NO<sub>3</sub> and NH<sub>4</sub>). Table 3.14 shows a significant effect of two of these groups, namely the seawater-derived and the anthropogenically-derived ones. In Figure 3.19, the effect of the deposition terms is 'isolated' from the other terms (that have a much stronger effect) by declaring these 'other' terms as covariables. This has no influence on the percentages explained variance, but it has an influence on the biplot, which now represents the effect of the deposition variables only, after accounting for the effect of the other ones.

The hypothesis that the first axis in Figure 3.19 represents a real effect of deposition is supported by the analysis of the Ellenberg indicator values in Table 3.17. On this axis, deposition of  $NO_3$ has a negative score, and deposition of Na has a positive score. The seawater-derived ions are correlated with many other factors that have a relation with distance to the coast (e.g., climate), whose cumulative effect on the species is reflected in Ellenberg's continentality indicator. Therefore, the first axis would be expected to be negatively correlated with the continentality indicator if it represents a real effect of Na or other seawater-related factors. Such a negative relation is found indeed (Table 3.17). The anthropogenically-derived ions are correlated with the general level of human activity, which tends to increase nutrient availability, either through atmospheric deposition or though ecosystem disturbance. Therefore, the first axis would be expected to be negatively correlated with the nutrient indicator if it represents a real effect of  $NO_3$ or other anthropogenic factors. This relation is found indeed (Table 3.17).

There are no indications that the second axis in Figure 3.19 also represents a 'real' effect of deposition. Instead, this axis seems to represent an effect of water availability, as shown by the relation with Ellenberg's humidity indicator and the species' ecology. As there are no

measurements of groundwater table or other indicators for water availability, this hypothesis cannot be tested on the basis of the present data.

The results indicate that the influence of bulk deposition on the composition of the ground vegetation is small. However, some important effects of deposition could be hidden within the variation explained by the traditional stand and site factors. First of all, precipitation is correlated with bulk deposition of nitrogen and also tree species may include deposition effects since dry deposition is generally higher on conifers, specifically spruce, than on deciduous trees. Furthermore, there is a relationship between the actual soil pH and historic acid deposition on the plot that could hide an effect of acidification, even though this influence is not so obvious from the results of this study (limited correlation between this variable and atmospheric deposition; see Table 3.11). Finally, introducing "country" as a variable could hide valuable information on deposition effects, as stated before. In summary, the real deposition effect is likely to be larger than the direct explained variance due to bulk deposition.

Despite the aspects mentioned above, it may seem amazing that no clearer impacts can be demonstrated of decades of acid and nitrogen deposition in terms of absence or presence of species, especially as the effects of nitrogen and acidity have been claimed to be large in many high deposition areas included in this study (cf. Van Dobben et al., 1999). It should be stressed, however, that this conclusion is only based on the spatial pattern of both vegetation and predictors. In interpreting the low percentage variance explained by the deposition terms, not only the correlation of deposition with the stand and site factors should be kept in mind, but also that the total variance in the present dataset is extremely large, as it covers forests of all climate zones and soil types over Europe. Therefore the effect of climate and soil is far larger than the effect of deposition. Rather, the effect of deposition should be considered as a weak 'signal' that is to be separated from large amount of 'background noise' caused by the traditional factors. In this view, it is already a clear signal that a significant effect of deposition is found anyway. Only in repeated measurements the 'background noise' is cancelled out, and the effect of (a change in) deposition can be determined with more certainty.

It may still be possible that there is a strong effect of deposition on vegetation in the temporal domain, for example that nitrogen-demanding species show a strong increase in places where deposition is high. However, the determination of such relations is outside the scope of the present study, and will only become possible when sufficient repetitive measurements are available. By continuation of this survey, ICP Forests will have data not only on distribution but also on any change in plant community over the past 5 years. This will allow a study on impacts of environmental factors on temporal changes, probably within 2 - 4 years.

## Species numbers and Simpson index

Just like the species composition, the species numbers and Simpson index seem to be mainly determined by the 'traditional' factors soil, climate and tree species. Here too the relations are in agreement with phytosociological literature (e.g. Braun-Blanquet, 1964) and national vegetation surveys (e.g., Stortelder et al., 1999). 'Rich' soils (i.e., high pH, high base saturation and high availability of base cations), southern climates and oak forests generally have a high diversity. In interpreting the figures for the Simpson index and the number of species it should be kept in mind that these two are rather strongly correlated (r = 0.61). Again the influence of deposition seems to be very limited, although significant effects of both seawater-derived, soil-derived and anthropogenically-derived ions are found (Table 3.18 and 3.19). However, the best-fitting models

show no effect of anthropogenically-derived ions (on the number of species), or no effect of deposition at all (on the Simpson index). In the models that do suggest a significant effect of deposition, the seawater-related ions Cl and Mg have a negative effect on biodiversity, and the soil-derived ones have a positive effect. There is no apparent explanation for these relations. The anthopogenically-derived ions have no consistent effect on biodiversity: there is a positive effect of NH<sub>4</sub> on the Simpson index in one of the models, while there is a negative effect of NO<sub>3</sub> on the species number in one other model. Also these effect have no apparent explanations. In general the effect of human activity on species diversity on plot level can be both negative (by the extinction of rare species) and positive (by the introduction of new species e.g. weeds).

In the analysis of the species numbers and Simpson index the assumption was made that the simple species lists that are the basis of this analysis are little influenced by methodological differences and therefore no adjustments were made for the effect of the countries. Therefore the effect of predictors that vary on a regional scale may be more pronounced in this analysis compared to the CCA analysis. This may be an additional cause for the significant effect of some of the deposition terms that do not appear in the CCA models.

# 3.4.3 Uncertainties in relations between species composition and environmental factors

# Reliability of results in view of differences in sampling design and data collection

This is an observational study, which means that predictors may be confounded with other, possibly not measured, variables, leading to spurious effects. In the present project it has been attempted to include all measurable variables that influence ground vegetation, with the exception of light and groundwater table. For both observational studies and designed experiments the implicit hypothesis is made that the studied situations are representative for other, non-studied situations. It is sometimes attempted to accomplish representativity by taking observation sites at random or at regular intervals. Such a design creates the need for a huge number of observations in order to reach the necessary variation in predictor and target variables. Actually, this would imply that one needs to do all the measurements at e.g. the Level I plots (statistical design). In the present case it has been attempted to deliberately locate the plots at sites that are representative for each country's forests. This is a generally accepted procedure which strongly reduces the cost of the project. However, to draw any conclusions from these data the assumption has to be made that the plots are representative indeed. This will not be completely the case and the results are strictly spoken only true for the plots under consideration.

Other limitations of the derived relationships are due to:

- Lack of data on stand structure and the limitation of the ground vegetation species to vascular plants. The exclusion of bryophytes and lichens, which was needed because their incomplete assessment, does affect the results. For example, lichens are known to be much more sensitive to air pollution than vascular plants and relationships derived are thus only relevant for the considered plant species, and not for the total plant biodiversity.
- Differences in data assessment methods between countries, such as different plot sizes and the use of fenced and unfenced plots. These differences influence the data comparability in an unknown way, since there are no intercalibration programmes to quantify the differences. These programmes are also needed to check the quality and accuracy of the observations, which is presently unknown. The fact that 13% of the variation in species composition can be

explained by country illustrates that differences in data assessment methods do affect the results from the ground vegetation survey.

## Reliability of results in view of available knowledge

In general, the relation between ground vegetation and environmental factors as found in this study is not different from the relation as postulated in the phytosociological literature (Braun-Blanquet, 1964; Oberdorfer, 1977, 1978, 1983; Runge, 1986; Rodwell, 1990; Van der Werf, 1991; Stortelder, 1999). However, two factors that are generally considered important could not be included in the present study by lack of data. These factors are water availability and light climate.

The results of the present study show a strong agreement with the results of a comparable study in The Netherlands (van Dobben and de Vries, 2001). In the latter study too, soil chemistry and tree species were found to be the most important predictors for ground vegetation, while the effect of atmospheric deposition was statistically significant but very small. The percentages explained variance found in this study were slightly higher compared to the figures presented here (c. 30%), which can probably be explained from a greater uniformity of methods, a higher number of measured predictors in combination with a smaller number of plots, and the use of RDA (='canonical' PCA) instead of CCA. Of the explained variance, 16% was due to tree species, 9% to soil chemistry, 3% to water availability (precipitation and groundwater level) and 2% to atmospheric deposition. Of the latter 2% explained variance, 1% is due to Mg (seawater-derived) and 1% to SO<sub>4</sub> (both anthropogenical and seawater-derived). Also the negative effect of beech on species diversity was found in this study.

## Differences between multivariate analysis and univariate analysis

When comparing the results of the multivariate analysis with the species-by-species univariate analysis, one gets the impression that for the present type of data (i.e., with many predictors, most of which only have a weak effect on the species), the multivariate approach is both the most sensitive and the most reliable one. This was exactly the conclusion of a theoretical study by Van Dobben and Ter Braak (1993) who experimented with synthetic datasets with different degrees of variability. When the relation between dependant variables and predictors is strong, both methods yield approximately the same results. In the present study this is the case for pH, where both methods show a negative relation for species like Vaccimium myrtillus, Deschampsia flexuosa and Calluna vulgaris, and a positive relation for e.g. Hedera helix and Anemone nemorosa. However, when the relation is only weak, the univariate methods are hampered by the type I error, because an apparently significant effect will be detected anyway for each predictor in one of each 20 species when P=0.05 is taken as the significance limit. Furthermore, if a weak effect of a predictor is causing a slight shift in the abundance of a number of species, the type II error will also be larger in univariate methods. If the effect is too weak to be significant in a species-byspecies analysis, a significant effect may be detected if all species are considered together as in multivariate methods. In the present data the latter seems to be the case for all predictors given in Table 3.14 except pH. It should however be stressed at this point that multivariate methods (and especially the unimodal ones like CCA) yield less quantitative information than a species-byspecies analysis. The fitting of response curves is not possible in multivariate methods, and therefore their use in predictive studies is only limited.

# Uncertainties due to lack of data

Uncertainties in the derived relationships are, amongst others, due to lack of relevant data influencing the species composition of the ground vegetation, such as water availability and light regime. The only predictor that is related to water availability is precipitation, and this predictor is not included in the 'restricted' model plotted in Figures 3.16 - 3.18. However, an additional 'passive' analysis (results not shown) indicates a significant negative correlation between precipitation and the sample scores on the second axis of the 'restricted' model, while also the Ellenberg humidity indicator has a negative relation with this axis. As the second axis is mainly representing an altitudinal gradient, with the higher altitudes at the axis' lower end, these relations are understandable in view of the generally higher precipitation received by mountain forest as compared to lowland forest. Apparently water availability is one of the factors that determine species composition of ground vegetation, but its effect is probably underestimated in the present study because no direct predictors (e.g. groundwater level) are available.

Also the light climate is lacking in the set of predictors that was used in this study. However, light climate is to a large extent determined by tree species, and it may be assumed that the effect of tree species is mainly or completely an effect of light climate. An indication for this is also the strong effect of beech as compared to the other tree species (beech is the first tree species selected in the forward selection procedure, and beech takes an extreme position relative to the third axis which is mainly representing the effect of tree species). This is understandable in view of the very dark light climate encountered in forests that have beech as the dominant tree species.

The results of this study allow the conclusion that the current programme can contribute to biodiversity issues in forests, since ground vegetation in combination with data on stand characteristics provide relevant information on this topic. Nevertheless, in view of the uncertainty of the results following from the sampling design and data collection, improvements are needed, such as (i) the collection of new data (e.g. inclusion of lichens and mosses at all plots), and (ii) the improvement of the comparability of data by writing a (sub) manual for the harmonised assessment of biodiversity parameters in the field and the set-up of intercalibration programmes to check the comparability of data. Ways for possible improvements are discussed below.

# **3.4.4 Future outlook**

The Ministerial Conference for the Protection of Forests in Europe (MCPFE), through its Conference process, has outlined the importance of Sustainable Forest Management, including the protection and enhancement of forest biological diversity. Recently, ICP Forests has therefore amended its mandate to include contributions, by means of the monitoring activities, to biodiversity assessment in forests. In collaboration with the European Commission, a "Biodiversity Working Group" has been formed (under the auspices of the Expert Panel on Ground Vegetation) to address the issue of forest biodiversity within the pan-European Monitoring Programme.

A range of different approaches exist to characterise forest biodiversity. The Biodiversity Working Group is now investigating the possibility of adopting the stand-scale structural approach, using the description of the forests stand as in indicator of forest biodiversity, in addition to species composition of the ground vegetation. The assumption behind this approach is that the range of habitat types, and thus the biodiversity potential, increases in more structurally diverse forest stands in terms of the presence or absence of vertical and horizontal layers. Structural descriptions are complimented with the addition of other information such as stand age and management regime, number of tree species and whether tree species are native or introduced to the region. The importance of forest deadwood to biodiversity is now widely recognised and a measurement of this too may be added as an assessment parameter. In addition to environmental factors, the occurrence and distribution of plant communities recorded at the plots may also be related with these structural indicators.

A list of proposed assessment parameters is presented in Table 3.20, with a rationale on how they might be used for the purposes of biodiversity assessment in forests. Some parameters are new to the monitoring programme, to enhance any assessment of forest biodiversity.

Parameter	Availability	Rationale/relevance
Tree species	Information on all tree species on the plot.	Mixed forest types are thought to be more diverse than monocultures. Tree species mixture also provides information on the horizontal structure of the stand.
Exotic vs	Information on tree species. From this, it	Native trees are thought to be associated with a greater
indigenous tree species	may be possible to describe whether a tree is native or exotic to the region.	biodiversity potential.
Stand age	Available in 20 years intervals. Ecosystem age may not however, be available.	Older forests are thought generally to be more diverse.
Vertical	Tree species mixture. From this it might be	More vertical layers in a forest stand may again be
structure	possible to describe the vertical structure of	associated with greater habitat diversity.
(number of	the forest stand, i.e. the number of layers. If	e ,
levels in stand)	not, it may be worth recording in the field with a standardised approach.	
Horizontal	Information on horizontal stand structure	A high standard deviation in dbh measurements implies
structure	(dbh measurements) from the forest growth survey.	a diverse horizontal structure.
Presence of	Can be obtained from the growth data,	The presence of large trees in a forest stand provides an
large old trees	although a precise definition of a "large" tree is required. Presence of those trees in the stand could be recorded.	important habitat for both fauna and epiphytic flora, including fungi.
Ground Vegetation	Species composition of the ground vegetation component at most plots: species number, the presence of rare plant species and invasive plant species (e.g. nitrophilous grasses) and cover abundance data of the plant species.	This data could also then be examined in relation to the deposition data at the Level II plots and the stand structure data to determine the effects of air pollution on forest biodiversity. A second ground vegetation survey is foreseen in the near future.
Lichens,	Available at part of the plots, since the	This component of the ground vegetation may account
bryophytes and epiphytic flora	recording of bryophytes and lichens, both ground dwelling and epiphytic, is not mandatory in the ground vegetation survey. It is recommended that the recording of bryophytes and lichens (including epiphytes) be made a mandatory parameter before the next ground vegetation survey of the Monitoring Programme foreseen in 2002 / 2003.	for a significant portion of forest biodiversity, particularly in plantation forests. They are also one of the first forest communities to demonstrate a response to air pollution and some species may be used as indicators of air quality.
Canopy closure	Hardly or not available, since canopy closure is not a mandatory parameter. Methods are available to assess canopy closure in the field but require standardisation before being introduced.	The abundance of ground flora (and epiphytes) depends heavily on the quantity and quality of light intercepting the forest floor and therefore this parameter becomes crucial for any multivariate analysis of plant distribution.

 Table3.20
 Proposed assessment parameters in view of their availability and their relevance

Parameter	Availability	Rationale/relevance
Natural	Information on natural regeneration may	The occurrence of natural regeneration in a stand is an
regeneration	already be available from the ground	important indicator for future biodiversity. The
	vegetation data set or could be recorded in	regeneration of native tree species is thought to be most
	the field.	important for biodiversity.
Stand history	Limited available. This information should	Stand or site history is a very valuable addition for
and management	be recorded where available for the plots.	biodiversity assessment in forests, e.g. whether the
regime (legal status)	Information on forest ownership may also be useful.	forest is a first rotation vs. Semi-natural or old growth forest. The management regime is one of the most important factors influencing forest biodiversity. Examples are whether the forest is managed for timber production, or recreational and social purposes, or whether the forest enjoys protected status.
Forest deadwood	Not available, since the occurrence of forest deadwood is not routinely recorded in the Monitoring Programme. Assessments should record the presence or absence of forest deadwood (quantity of the resource) and the decomposition status of the wood (quality of the resource). Differentiation should also be made between standing and lying deadwood. Again, such assessments would need to be carried out in a standardised manner.	Forest deadwood, both on the forest floor and standing dead is important in providing habitat, shelter and nourishment for a variety of organisms. Therefore, any assessment of forest biodiversity would be complimented greatly by inclusion of a deadwood assessment.
Litterfall	Surveys of forest litterfall are carried out at a selected number of plots	Provides valuable information on nutrient inputs to the forest floor, which in turn have a strong influence on the soil biotic community. Changes monitored in these inputs over time may be related to changes in the forest floor communities, both floral, faunal and indeed fungal.
Habitat	No information on the occurrence of	Where these habitat types and others occur they should
information	different habitat types, e.g. the presence of standing or running water, the occurrence of open spaces in the stand etc.	be recorded according to a standardised reporting form.
Forest	Stratification into broad forest types, e.g.	The incorporation of these forest types into the stand
stratification	following the BEAR project could be done	description process would be most useful.
Forest pests and	Recently, ICP Forests and the European	Results of any such survey could be useful to
diseases	Commission have set up a Working Group on Biotic Damage Assessment.	understand the occurrence and distribution of specific invertebrate and fungal groups in forests.
Natural	Assessed but not recorded at a European	The importance of natural disturbance events in forests
disturbance	scale. Disturbance events include events	on forest biodiversity is widely recognised (e.g. canopy
events	such as forest fire, windblow and snow damage.	opening following windblow, nutrient release and loss of habitat following forest fire, etc.).
Remote sensing	Remote sensing data exists for a selection of plots and this may be used to describe the landscape diversity where applicable.	Remote sensing data may be used to describe the extent of the forest landscape diversity, including estimates of forest cover continuity, forest connectivity and forest fragmentation.

This structural approach of forest biodiversity characterisation assumes that specific structural characteristics of the forest may be used as an indicator of the biological diversity of the forest stand. This approach has been well elaborated recently by the BEAR project ("Indictors for monitoring and evaluation of forest biodiversity in Europe", Larsson 2001). However, the relation between biodiversity and structural parameters is based on circumstantial evidence rather than on hard data. Therefore the extensive data collected by ICP Forest create a unique opportunity to validate such relationships, at least for the ground vegetation. This would create the possibility to upscale biodiversity assessment to a finer geographical resolution, using less detailed forest inventory data. In this way, comparatively little additional effort would be needed to make the ICP Forests and European Commission pan-European Monitoring Programme an effective tool to assess the biodiversity of the forests of Europe. Such an extension of the Programme's objectives

might create a need for the collection of additional data, e.g. the ones related to forest structure summarised in Table 6.1. However, although in this way the Programme could significantly contribute to biodiversity issues, biodiversity measures will probably remain limited to the floral component of the ecosystem. The collection of data on fauna (with the possible exception of a few groups e.g. plague insects) is not feasible within the given framework. Only the expansion of the vegetation to cryptogamic species, including epiphytes, seems a useful addition at this point. Therefore, with the addition of comparatively few new assessment parameters, the pan-European Monitoring Programme may be used as a cost effective multifaceted approach for describing forest biodiversity at a European scale. In this context, the Biodiversity Working Group is preparing a submanual for the harmonised assessment of such parameters in the field, as proposed at the 18th Task Force of ICP Forests in May 2002.

# **3.4.5** Conclusions

In summary, the results obtained allow the following main conclusions:

## Geographical variation of biodiversity indices and Ellenberg indicator values

The evaluation of ground vegetation data in terms of species numbers, species diversity and Ellenberg indicator values allow the following conclusions:

- Species numbers show a slight North-South gradient with increasing species numbers in the Mediterranean areas compared to the boreal forests. This is in agreement with common knowledge.
- The Simpson species diversity index shows large, rather random differences in species diversity between plots within a country, in which only a very slight North-South gradient can be detected.
- Ellenberg indicator values for temperature and soil acidity, that could be derived from the plant species composition data, show a clear north- south gradient. In line with common knowledge, the Ellenberg values indicate low pH and temperature in the north (cold acid circumstances) to high temperature and pH in the south (warmer more alkaline circumstances).

# Relationships between the occurrence probability of species and environmental factors

Derived relationships between the occurrence probability of individual species and environmental factors for 332 different species allow the following conclusions:

- The median explained deviance of the models constructed for all individual species is about 30%, with the deviation varying mostly between 10 and 70%.
- For a limited number of species (36), there is a significant relationship between the occurrence probability and soil pH, with most species favouring alkaline conditions and few species being more prominent under acid conditions. The Ellenberg indicator values for pH, based on species composition are significantly related to measured soil pH values.
- A relationship between occurrence probability and atmospheric nitrogen deposition was found for a few individual species, some favouring nitrogen rich and some nitrogen poor circumstances.

# Relationships between the species composition of ground vegetation and environmental factors

Derived relationships between the species composition of ground vegetation and environmental factors, related to soil, tree species, climate and atmospheric deposition, allow the following conclusions:

- Approximately 40% of the variation in species numbers can be explained by environmental factors, whereas the explanation of the Simpson index is approximately 15%. The pH in the organic layer explains most of the variation, followed by tree species, soil factors related to nutrient availability, climate and atmospheric deposition.
- Approximately 15%-21% of the variation in the abundance of the various species occurring in the ground vegetation could be explained by the included environmental factors, depending on either the use of bulk depositon or throughfall. As with the species numbers and the Simpson index, the explained variance is mainly due to the soil acidity, tree species and climate in terms of precipitation and temperature, which contribute in approximately equal amounts to the fit of the model. In case of bulk deposition chemistry only a very small portion of the explained variance (0.4% out of 14.5% explained variance) is due to NO<sub>3</sub> deposition, being of anthropogenic origin. In case of throughfall, the explained variance increases to 1.2% out of a total explanation of 20.7%.
- The explained variance increases by 13% when country is included as an explicit predictor, but this only illustrates that part of the variation can be explained by differences in data assessment methods.

# Impacts of atmospheric deposition in view of the used approach

The results do show an effect of deposition, but it accounts for only a minor part of the variation, whereas most of the explained variation is due to stand and site factors, such as precipitation, tree species and soil pH. This conclusion must, however, be considered with care for the following reasons:

- Precipitation is correlated with bulk deposition of nitrogen and tree species with dry deposition (dry deposition is generally higher on conifers than on deciduous trees).
- There is a relationship between the actual soil pH and historic acid deposition on the plot that could hide an effect of atmospheric deposition
- Introducing "country" as a variable could hide valuable information on deposition effects.
- The results of this study are based on the spatial patterns of both vegetation and environmental factors in which the total variance is extremely large, as it covers forests of all climate zones and soil types over Europe. Only in repeated measurements the 'background noise' is cancelled out, and the effect of (a change in) deposition can be determined with more certainty.

In summary, the results have pin-pointed the methodological problems to relate stress and response and will hopefully catalyse the process of harmonisation of methods among the member states to solve this problem. Future work should focus on functional relationships between ground flora species composition and environmental and deposition variables measured in other sub-programmes. Within 2-4 years data on temporal changes in species composition are available. This will allow a more functional approach.

# 4 Critical loads and present deposition thresholds for nitrogen and acidity and their exceedances

# **4.1 Introduction**

# The critical load concept and its use in policy making

In the terminology of air pollution impacts, critical levels and critical loads refer to the concentration levels and deposition loads of air pollutants ( $SO_2$ ,  $NO_x$ ,  $NH_3$  and  $O_3$ ), respectively, below which no adverse (direct) effects are expected. The concept of critical levels and loads is based on the concept of thresholds. Woodwell (1976) defined an ecological threshold as the "maximum exposure (to toxins) that has no effect" and this concept was later applied to atmospheric pollution (Gorham, 1976; EPRI, 1991). In this chapter, we limit ourselves to a comparison of deposition loads from the atmosphere onto forests with critical loads. A comparison of the air quality with critical concentration levels for e.g. ozone or S and N compounds in the atmosphere was not yet possible because of lack of data.

The concept of critical loads and levels has been developed further in Europe during the last 15 years in the context of the work under the 1979 Convention on Long-range Transboundary Air Pollution (LRTAP). It strongly influenced international agreements in the nineties. The earlier protocols to the Convention (1985 on sulphur, 1988 on nitrogen oxides, 1991 on volatile organic compounds) were agreements based on a stand-still or flat-rate reductions of emission. The 1994 Sulphur Protocol was the first to consider ecosystem vulnerability in terms of critical loads for the formulation of reduction requirements. The assessment of critical loads is the core of the work of the ICP on Mapping and Modelling, established in 1989, under the Working Group on Effects (WGE) of the Executive Body of the LRTAP Convention. Under this ICP, critical load data from individual countries are collected, collated and mapped by the Coordination Center for Effects. Critical loads have thus been calculated by Austria, Belarus, Belgium, Bulgaria, Croatia, Czech Republic, Denmark, Estonia, Finland, France, Germany, Hungary, Ireland, Italy, Netherlands, Norway, Poland, Republic of Moldova, Russian Federation, Slovakia, Spain, Sweden, Switzerland and the United Kingdom (see Posch et al., 1999). These critical loads are provided to the relevant UNECE bodies under the LRTAP Convention as well as the European Commission to formulate and negotiate emission reduction strategies in Europe.

# Scientific discussions on the topic

The scientific discussion on critical loads (depositions) started at a workshop organised by the Nordic Council of Ministers (NMR) in 1986 in Sundvollen (Norway). It provided for the first time, estimates of critical loads of sulphur and nitrogen for forest soils, groundwater, and surface waters (Nilsson 1986). The first workshop on critical loads held under the auspices of the United Nations Economic Commission for Europe (UNECE), which provides the permanent secretariat for the LRTAP Convention, was organised in 1988 by the NMR at Skokloster (Sweden). At this workshop, the still-valid definition of a critical load as "the quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" was agreed upon (Nilsson and Grennfelt, 1988).

The basis of the mapping of critical loads in the ECE countries was laid at a UNECE workshop held in 1989 in Bad Harzburg (Germany). This resulted in a manual for mapping critical levels and loads, which has been updated periodically (UNECE, 1996). Furthermore, in a workshop on critical loads of nitrogen organised by the NMR in Lökeberg (Sweden) in 1992 recommendations for deriving critical loads of nitrogen and their exceedances were elaborated (Grennfelt and Thörnelöf, 1992). Remaining open questions were discussed at a UNECE workshop in Grange-over-Sands (England) in 1994, organised by the UK Department of Environment (Hornung et al., 1995). More information on the critical load concept and related scientific discussions is given in Posch and De Vries (1999).

# Differences between critical loads and present deposition thresholds

Critical loads for nitrogen and acidity are mostly calculated with steady-state soil models based on a simple mass balance approach. These models calculate deposition loads that avoid the violation of a soil chemical criterion in a steady-state situation. The major premise of the calculation is that it assumes a steady-state situation to derive a long-term acceptable or critical load. Processes that play a role on a finite time scale only, such as cation exchange and sulphate adsorption, are not included. This implies that critical loads are relevant to assess the ultimate emission reductions, but an excess of those loads does not necessarily imply that the forest ecosystem is at risk yet. To gain insight in the relationship in ecosystem risk and atmospheric deposition, we need to know the deposition threshold that violates a soil chemical criterion at present. Such thresholds are denoted as present deposition thresholds, being the deposition loads that lead to concentrations of nitrogen and acidity in soil solution that are equal to critical limits at present (not in a steady-state situation). Present deposition thresholds can thus become very high, e.g. in well buffered clay soils with a high base saturation. The value presents the amount of acidity that is needed to bring the soil directly to a state where it violates a critical limit, e.g. for aluminium.

# Role of ICP Forests in deriving critical loads

Calculations of critical loads have generally been based on estimated average data on tree uptake, soil weathering and nitrogen retention, hardly including actually measured data at Intensive Monitoring plots. Up to now, such calculations have only been made for the German Intensive Monitoring plots (Becker et al., 2000; Becker and Gehrmann, 2001; Becker and Nagel, 2001). This report presents a European wide assessment of critical loads for Intensive Monitoring plots in comparison to present loads. As such, this work under ICP forests has a clear contribution to the work of the ICP Mapping and Modelling, in that it provides high quality data for the ultimate European wide assessment. Furthermore, it also includes results for present deposition thresholds for plots where all needed data on deposition, meteorology, forest growth and soil and soil solution chemistry were available. The latter result is relevant when assessing relationships between present loads and impacts on forests

# Contents of this chapter

This chapter first presents the methods (models, critical limits and input data) that are needed to calculate critical loads and present deposition thresholds (Section 4.2). Results are described in Section 4.3. This includes critical loads and present deposition thresholds for nitrogen and acidity in comparison to present loads, distinguishing between impacts on the soil, ground vegetation and trees. Finally, a discussion of the results and conclusions are presented in Section 4.4.

# 4.2 Methods

In this section, first the locations are described for which critical loads or present deposition thresholds were calculated, in combination with present loads (4.2.1). This section is followed by a general description of approaches that can be used to calculate critical loads, focusing on the model approach that is used in this study (Section 4.2.2). The critical limits, soil models and input data that were used to calculate critical loads and present deposition thresholds of nitrogen and acidity are presented and discussed in Section 4.2.3 and Section 4.2.4, respectively.

# 4.2.1 Locations

Critical loads of nitrogen (N), being the sum of NO<sub>3</sub> and NH<sub>4</sub>, and of acidity, defined as the sum of S and N (see section 4.2.4.2) were calculated for all Intensive Monitoring plots for which annual water fluxes (precipitation excess) were available. This included 245 plots for the period up to 1998. Actually, data on both rainfall and throughfall were available for 309 plots, but at 64 plots, hydrological fluxes could not be successfully calculated due to inconsistencies in the provided data, as described in the previous technical report (De Vries et al., 2001). Present atmospheric deposition of nitrogen and acidity on forest stands up to 1999 could be calculated for more than 300 Intensive Monitoring plots where both bulk deposition and throughfall deposition of sulphate (SO<sub>4</sub>), nitrate (NO<sub>3</sub>) and ammonium (NH<sub>4</sub>) was measured, but the calculations were limited to the 245 plots for which critical loads could be calculated. Ultimately, a comparison of present and critical loads of nitrogen and acidity could be made at 234 and 226 plots, respectively, due to lacking data on other aspects than water fluxes. The 234 plots for which both present loads and critical loads could be calculated were located in 13 different countries, mostly in Central and Western Europe (Fig 4.1).

Figure 4.1 also shows the plots for which present deposition thresholds were calculated. This included the plots with information on deposition and leaching and thus on retention or release of nitrogen, sulphur and base cations, based on element budgets. Such budgets were only available for 121 plots, due to the limited availability of soil solution chemistry data, as described in De Vries et al. (2001). These plots were located in Belgium, France, Denmark, Germany, UK, Ireland, Norway, Sweden, Finland and Austria (Fig. 4.1).

# 4.2.2 Approaches to derive critical loads for forest ecosystems

With respect to the assessment of critical loads, a major distinction can be made between an empirical approach and a model-based approach (Figure 4.2). More specifically, a distinction can be made in an (De Vries and Latour, 1995):

- Empirical approach, in which critical loads are derived from observed relationships between atmospheric deposition and effects on "specified sensitive elements" within an ecosystem (ecosystem effects) by correlative or experimental research.
- Soil model-based approach, in which critical loads are derived with soil models, using environmental quality criteria or critical limits for element concentrations or element ratios in the soil solution. Such limits are based on dose response relationships between these element concentrations or ratios and the ecosystem status, derived from laboratory or field research.
- Integrated model-based approach, in which critical loads are derived with integrated soil-vegetation models, which include biotic interactions, on the basis of criteria for all parts of the forest ecosystem.

In this study, critical loads were calculated with a soil model-based approach for approximately 230 Intensive Monitoring plots , where data on deposition, meteorology, forest growth and soil chemistry were available. Two different types of critical loads were calculated, requiring either:

- 1. No further net accumulation of nitrogen or loss of exchangeable base cations in the forests soil (stand-still principle) or
- 2. Concentrations of nitrogen or acidity that stay below critical limits in soil solution (effectbased principle).

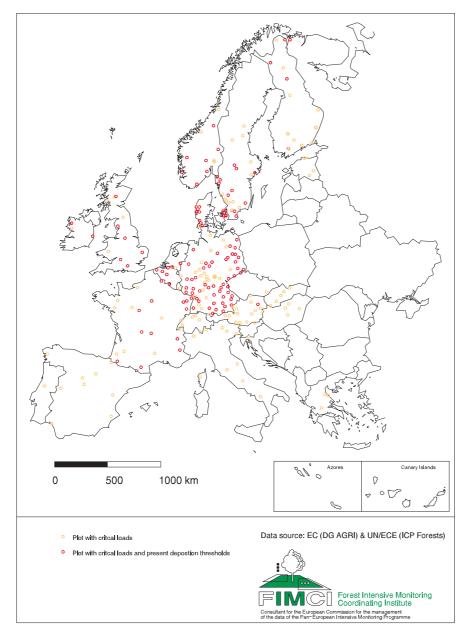
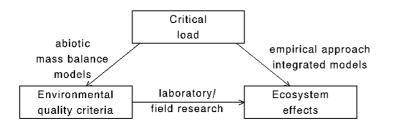
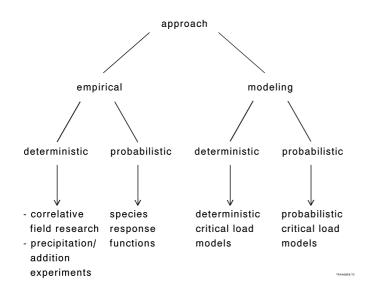


Figure 4.1 Geographical location of plots for which critical loads or present deposition thresholds were calculated



*Figure 4.2 Methods to derive critical loads (each arrow indicates a relationship than can be assessed by correlative research, process research and/or model research).* 

Below, a description is given of empirical and soil model-based approaches, including a further distinction in deterministic and probabilistic methods (see Figure 4.3). Generally available results of empirical approaches are presented in Section 4.2.2.1, since those data are relevant for evaluating results of the soil models used in this study (section 4.2.2.2). In the section on empirical probabilistic approaches, results of integrated models including empirical aspects are also presented. Integrated models are, however, not further discussed here, but an evaluation of those models is given in Section 4.4.



*Figure 4.3 Overview of possible empirical and model based deterministic and probabilistic approaches to derive critical loads.* 

#### 4.2.2.1 Empirical approaches: data to evaluate results of model approaches

#### Deterministic approaches

Deterministic empirical approaches have been used to derive critical nitrogen loads in view of adverse impacts on plant biodiversity, since nitrogen has a dominating influence on the species diversity of terrestrial vegetation. Empirical critical N loads for terrestrial ecosystems, related to changes in vegetation and fauna, are based on an extensive but inhomogeneous summary of field studies and large-scale laboratory (greenhouse) experiments (cf. Bobbink et al., 1992, 1995, 1998). This includes: (i) precipitation experiments with NH<sub>4</sub> on small-scale heathland in a

greenhouse and (ii) correlative field studies between N deposition and species diversity, using present geographic differences or historical data on N deposition and species decline (cf. De Vries and Latour, 1995). Critical N loads have, for example, been estimated by comparing the N deposition on grass dominated and heather dominated heathlands (e.g. Liljelund and Torstensson, 1988) or by experimental investigation of the biomass development of grasses in heathlands as a function of N input (e.g. Roelofs, 1986).

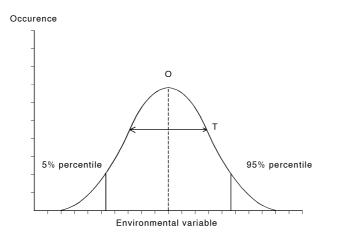
An overview of empirical data for critical N loads on forest ecosystems, summarised by Bobbink et al. (1998) is given in Table 4.1. The values are related to impacts of nutrient imbalances on trees and to vegetation changes for the ground vegetation. In general, average critical loads related to impacts on trees are approximately 20 kg N.ha<sup>-1</sup>.yr<sup>-1</sup>, varying between 10-30 kg N.ha<sup>-1</sup>.yr<sup>-1</sup>. Critical loads related to impacts on the species composition of the ground vegetation are generally lower, varying between 7-20 kg N.ha<sup>-1</sup>.yr<sup>-1</sup>, with an average value of approximately 14 kg N.ha<sup>-1</sup>.yr<sup>-1</sup> (Table 4.1).

Table 4.1Summary of empirical critical loads of N (in kg N.ha<sup>-1</sup>.yr<sup>-1</sup>) for forest ecosystems, related to forest<br/>condition and species composition of ground vegetation. (## reliable; # quite reliable; (#) best guess :<br/>after Bobbink et al., 1998).

after Bobbink et al., 1998	9.	
Forest ecosystem	Critical load kg N.ha <sup>-1</sup> .yr <sup>-1</sup>	Indication
Forests: trees		
Acidic coniferous trees	10-15 #	Nutrient imbalance (low nitrification)
	20-30 #	Nutrient imbalance (moderate – high nitrification rate)
Acidic deciduous trees forests	15-20 #	Nutrient imbalance; increased shoot-root ratio
Forests: ground vegetation		
Acidic managed coniferous forests	7-20 ##	Changes in ground flora and mycorrhizae; increased N leaching
Acidic managed deciduous forests	10-20 (#)	Changes in ground flora and mycorrhizae
Acidic unmanaged forests	7-15 (#)	Changes in ground flora; increased N leaching
Calcareous forests	15-20 (#)	Changes in ground flora

# **Probabilistic** approaches

The drawback of deterministic empirical critical (nitrogen) loads is that they do not give equal protection percentages to the species occurring in different ecosystems. The concept of risk assessment may be used in this perspective as an alternative, because it provides a framework to achieve more standardisation in the assessment of protection levels for different environmental problems (e.g. Latour et al., 1994). Most progress in assessing and quantifying ecological risks has been made in the field of toxicological stress. There the maximum tolerable concentration (MTC) is chosen as the environmental concentration of a compound (e.g. heavy metal) at which (theoretically) 95% of the species are fully protected. MTCs are calculated by extrapolation of "No Observed Effects Concentrations" (NOEC levels) for single-species to an ecosystem, mostly assuming a log-logistic, or log-normal distribution of species sensitivities (Slooff, 1992). At the species level the risks are assessed based on the species-response function, which describes the occurrence probability of a species as a function of an environmental variable. The speciesresponse function can be characterised by its optimum (O) and standard deviation. Latour et al. (1994) used the 5-th and 95-th percentiles of the species-response curves as NOEC-like measures for the risk at the species level (Figure 4.4). The 5-th percentile corresponds to a reduced occurrence probability due to "limitation", the 95-th percentile due to "intoxication". Species are considered protected between the 5-th and 95-th percentile of a given environmental variable.



*Figure 4.4 Probability of the occurrence of a species as a function of an environmental variable (species-response function). Between the 5-th and 95-th percentile, species are considered protected.* 

An example of a probabilistic empirical model for deriving critical N loads is the model MOVE (multiple-stress model for vegetation; Latour and Reiling, 1993). MOVE predicts the occurrence probability of about 700 species as a function of three abiotic soil factors: soil acidity, nitrogen availability and soil moisture. With regression statistics the occurrence probability of a species can be calculated for each combination of the soil factors or for each factor separately (species-response function). Since combined samples of vegetation and environmental variables are rare, the indication values of plant species by Ellenberg (1991a) are used to assess the abiotic soil conditions. Combined samples of vegetation with environmental variables are used to calibrate Ellenberg indicator values with quantitative values of the abiotic soil factors.

Latour et al. (1994) used the model MOVE to derive critical N loads for fertilised grasslands in the Netherlands, based on NOECs for 275 species. More recently, Van Hinsberg and Kros (1999) derived critical loads for nitrogen related to species diversity for the most common terrestrial ecosystems in the Netherlands with the probabilistic integrated SMART-MOVE model. A comparison of their results with empirical data for forest ecosystems is given in Table 4.2. In general, the results with SMART-MOVE were within the range of empirical critical N loads for acidic forests, but for calcareous forests, the results obtained with SMART-MOVE were clearly higher.

Table 4.2 Comparison of empirical data for critical N loads related to terrestrial ecosystems derived by Bobbink et al. (1998) and the probabilistic integrated soil-vegetation model SMART/MOVE using a protection percentage of 80% (after Albers et al., 2001).

Ecosystem	Critical load (kg N	.ha <sup>-1</sup> .yr <sup>-1</sup> )
	Empirical data	SMART/MOVE
Acidic, managed coniferous forests	7-20	18 (poor sandy soils)
Acidic, managed deciduous forests	10-20	18 (poor sandy soils)
		22 (rich sandy soils)
Calcareous forests	15-20	17 (loess soils)
		33 (marine clay soils)
		38 (calcareous dunes)

## 4.2.2.2 Modelling approaches: used to calculated critical loads

In this study, models are used to quantify critical loads at the Intensive Monitoring plots. A major challenge in this context is to transform empirical findings and conceptual insight into robust ecosystem models, requiring a minimum of data, while still accounting for the most important factors controlling critical loads. In general, critical loads can be calculated either by steady-state models or by dynamic models with different degrees of complexity, as discussed below. The models used are all deterministic, but a probabilistic approach is possible by combining them with Monte Carlo analyses, including ranges in input data instead of fixed values. These models allow the calculation of critical loads on the basis of critical limits for the soil or soil solution.

# Critical limits

In a model based approach, critical loads are derived with models using critical limits for element concentrations or element ratios in the ecosystem, based on dose response relationships between these critical limits and the ecosystem status. Using this approach, a critical load of nitrogen, or acidity equals the deposition resulting in a concentration in a compartment (e.g. soil, groundwater, plant, etc.) in a steady-state situation that does not exceed a critical limit, thus preventing 'significant harmful effects on specified sensitive elements of the environment'. Consequently, the selection of critical limits is a step of major importance in deriving a critical load.

In defining a critical load, one aims at long-term protection of the ecosystem. Critical loads of nitrogen and acidity are derived by setting a limit to the leaching of nitrogen, acidity or metals. In this context, protection can be defined as the situation where:

- No further net accumulation of nitrogen or loss of exchangeable base cations or readily available aluminium occurs (stand-still approach). This implies that the present value for the concentration of nitrogen, metals, exchangeable base cations or Al compounds in the soil is considered the critical limit, above which no further increase or decrease is accepted. In Table 4.3 the criteria used are defined. These values are combined with process descriptions to relate them to dissolved concentrations to calculate critical leaching rates.
- Concentrations of nitrogen or acidity stay below critical limits in defined compartments in a steady-state situation (effect-based approach). The critical limits are based on adverse effects on (parts of) the ecosystem, such as the soil solution or plants, and the nitrogen, acidity or metal concentrations should stay below those limits.

To date mostly soil chemical criteria (e.g. critical nitrate, aluminium or metal concentrations or critical aluminium to base cation ratios) have been used to derive critical loads with simple steady-state models. The largest uncertainty in these calculations remains the relation between the critical limits and the "harmful effects". In Table 4.3 an overview is given of relevant critical limits for nitrogen and acidity used in the calculations. More information is given in Section 4.2.3.1 and 4.2.4.1.

Critical limits			
Nitrogen	Acidity		
$\Delta N=0$ ; N in solution	$\Delta BS=0$ and/or $\Delta Al_{ox}=0$		
N in solution = $0.2 \text{ mol}_{c}.\text{m}^{-3}$	-		
N in solution is related to critical N content	Molar Al/(Ca+Mg+K) ratio = $0.8$ for conifers		
plant of 18 g.kg <sup>-1</sup> (app. 0.25 mol <sub>c</sub> .m <sup>-3</sup> )	and 1.6 for deciduous trees.		
	$\Delta N=0; N \text{ in solution}$ = 0.02 mol <sub>c</sub> .m <sup>-3</sup> N in solution = 0.2 mol <sub>c</sub> .m <sup>-3</sup>		

 Table 4.3
 Examples of critical limits used to derive critical loads

## Steady-state models

Critical loads for nitrogen and acidity are mostly calculated with a steady-state single-layer soil model based on a simple mass balance approach, called SMB model (Sverdrup and de Vries, 1994). This approach was also used in this study and is further elaborated in the Sections 4.2.3.2 and 4.2.4.2. It is also the standard for calculating critical loads for heavy metals, as described in Section 5.2.2.2. These models calculate deposition loads, which avoid the violation of a chosen soil chemical criterion in a steady-state situation. Therefore, processes with a finite time scale, such as cation exchange and sulphate adsorption, are not included. The models include a simple description of the inputs (deposition), outputs (leaching) and permanent sources and sinks of major ions within the rooting zone. For nitrogen, the processes involved are retention of N by net uptake, denitrification and net N immobilisation. For acidity, processes involved are net retention of N as above, net input of base cations (weathering minus net uptake) and release of Al from silicates and Al hydroxides. Ions involved include sulphate, nitrate, base cations and aluminium and their steady-state mass balances are combined with a charge balance of those ions in the soil leachate.

In steady-state one-layer soil models, the soil is considered a single homogeneous compartment with a depth equal to the root zone. This implies that internal soil processes (such as weathering and uptake) are evenly distributed over the soil profile, and all physico-chemical constants are assumed uniform in the whole profile. Furthermore the simplest possible hydrology is assumed: the annual water flux leaving the root zone equals the annual precipitation minus evapotranspiration. For both nitrogen and acidity, the SMB model has been and is widely used to produce maps of critical loads of S and N on a European scale (Posch et al., 1995, 1999).

It is also possible to use multi-layer models including element cycling (litterfall, mineralisation and uptake). An example for acidification is the MACAL model (De Vries et al., 1994b) and the PROFILE model (Sverdrup and Warfvinge, 1992), which has been applied in many countries (e.g. Sweden, Switzerland, Poland, Germany, Turkey, Czech Republic and Slovakia). Such models do have a high demand for input data. The steady-state one-layer soil models are primarily developed for applications on a large regional scale, for which data is scarce.

# Dynamic soil models

Dynamic soil models include the same processes as steady-state soil models, but simulate additional processes that play a role on a finite time scale. For nitrogen it includes a dynamic description of nitrogen retention or release. Additional processes in dynamic soil acidification models are cation exchange and sulphate adsorption. These models can also be used to calculate critical loads by running the model until a steady state is reached. By trial and error the (constant) deposition level is calculated that fulfils a chosen chemical criterion (see, e.g., Warfvinge and Sverdrup, 1995, where critical acid loads for Sweden are derived with the dynamic model SAFE). Critical loads calculated in this way are equal to those derived by steady-state models if the processes in both models are modelled in the same way. Dynamic soil models can furthermore be used to derive so-called target loads by considering a finite time period (e.g. one forest rotation) in which the system is allowed (or has to) reach a chemical criterion. Unlike the critical load, the present acidification status of the soil system and time-limited processes influence the target load. Finally, dynamic soil models can also be used to derive the time period before the system reaches a chosen soil chemical criterion for a given deposition scenario. Depending on the present soil status, the model thus calculates the time period before risk increases or before the system starts to recover. Dynamic models are most commonly used in this context (see Cosby et al., 1989; De Vries et al., 1994c). Some of the more widely used simple dynamic soil models are MAGIC

(Cosby et al., 1985) and SMART (De Vries et al., 1989). As with the steady-state models, there are also multi-layer dynamic models, such as SAFE (Warfvinge et al., 1993) to predict the dynamics in soil response at different depths.

# 4.2.3 Calculation of critical loads and present deposition thresholds for nitrogen

In this section, first an overview is presented of the impacts of nitrogen on forest ecosystems, followed by the critical limits derived and used in this study (Section 4.2.3.1). The background of the derivation of the simple mass balance model for nitrogen, used to calculate critical nitrogen loads in this study, is presented in Section 4.2.3.2. This section also includes the methods used to calculate of the critical N leaching rate and the present deposition threshold. The input data used in the calculations are described in Section 4.2.3.3.

# 4.2.3.1 Impacts of nitrogen and critical limits

# Impacts on forest ecosystems

The impact of N on an ecosystem depends on its nitrogen status, since N is a nutrient that may be either in short supply or in excess. Since the beginning of the eighties several authors (e.g. Ulrich et al., 1979; Ellenberg 1983, 1985 and 1991b; Nihlgård, 1985; Tamm, 1991; Gundersen, 1992) hypothesised that elevated inputs of N lead to vegetation changes as well as damage to trees. In systems with low N status, an elevated input of N will increase forest growth until a certain threshold level. Observations of increased tree growth of European forests (Kauppi et al., 1996; Spiecker et al., 1996) may be the effect of increased N inputs. Below the threshold level for reduced forest growth, however, changes in the ecosystem are observed, especially the species composition of the ground vegetation may gradually change towards more nitrophilic species (Ellenberg, 1985; Bobbink et al., 1995, 1998). A thorough review of the impacts of N inputs on the species diversity of terrestrial ecosystems in general, i.e. ombotrophic bogs and wetlands, heathlands, species-rich grasslands and forests, including empirical critical N loads related to vegetation changes, has been given in Bobbink et al. (1998).

In forested plots with a continuous high N input, essential resources other than N may periodically limit primary production, especially when the canopy reaches its maximum size and N utilisation efficiency decreases. The ecosystem then approaches 'N saturation' (Aber et al., 1989). A forest ecosystem leaching  $NO_3^-$  (or  $NH_4^+$ ) is saturated, but it may still respond to N additions and accumulate a considerable amount of N in the biomass. At the stage of 'N saturation' or 'N excess', the ecosystem may be destabilised by the interaction of a number of factors (Erisman and De Vries, 1999). These are:

- An increased water stress as a result of increased canopy size, increased shoot/root ratio, and loss of infection by mycorrhizae (De Visser, 1994);
- Root damage due to acidification caused by climatically controlled pulses of nitrification (Matzner, 1988);
- Nutrient deficiencies or nutrient imbalances (Nihlgård, 1985; Roelofs et al., 1985; Schulze, 1989), since the increase in canopy biomass causes an increased demand of base cation nutrients (Ca, Mg, K) whereas the uptake of these cations is reduced by increased dissolved levels of NH<sub>4</sub> and Al (Boxman and Van Dijk, 1988), a loss of mycorrhizae or root damage (Schulze, 1989).

- Accumulation of N in foliage (e.g. as amino acids), which may affect frost hardiness (Aronsson, 1980) and the intensity and frequency of insect and pathogenic pests (Popp et al., 1986; Roelofs et al., 1985).

In addition, the nitrate leaching to ground water, such that  $NO_3$  concentrations in (shallow) ground water exceed the current EC drinking water standard of 50 mg.l<sup>-1</sup> (e.g. Boumans and Beltman, 1991), has to be considered.

Experiments with decreased N deposition at N saturated sites, after building a roof below the canopy to prevent N inputs into the soil, showed an immediate decrease in nitrate leaching (Boxman et al., 1995; Bredemeier et al., 1995; Wright and Rasmussen, 1998). This shows that N saturation is a reversible process in chemical terms. In ecological terms, an improvement is to be expected as well, since several species returned during the period with low N input.

# Critical limits for forest ecosystems

A summary of critical limits for N or  $NO_3$  in plants or soil solution are given in Table 4.3. Below more information on the background of those limits is given

*Soil:* Crucial in the calculation is the choice of the critical limit determining the critical N leaching rate In deriving critical loads, the critical or acceptable N leaching rate has mostly been related to the loss of nitrogen from unpolluted ecosystems. The following ranges are often used (UNECE, 1996):

- 0.5-1 kg.ha<sup>-1</sup>.yr<sup>-1</sup> for managed coniferous forests
- 1-2 kg.ha<sup>-1</sup>.yr<sup>-1</sup> for Mediterranean forests
- 2-4 kg.ha<sup>-1</sup>.yr<sup>-1</sup> for temperate deciduous forests

Another option is to use a critical nitrogen concentration and multiply this value with the precipitation excess. This option was used in this study, using a critical limit of 0.02 mol.m<sup>-3</sup>, or 0.28 g.m<sup>-3</sup>, based on data from stream water of nearly unpolluted forested areas in Sweden (Rosén, 1990). With a precipitation excess of 200-800 mm.yr<sup>-1</sup>, this gives a leaching rate of 40-160 mol.ha<sup>-1</sup>.yr<sup>-1</sup> (0.5-2 kg.ha<sup>-1</sup>.yr<sup>-1</sup>), which is a common range of natural N losses from an ecosystem, as shown above.

In this study, we used the low natural leaching losses as a proxy to calculate the critical loads related to the stand-still principle, starting from a pristine situation. In this case, one should in principle use the present N leaching rates and not the natural leaching rates. The drawback of this approach is, however, that the present leaching rates can already be strongly elevated due to historic and present N deposition and might therefore not be acceptable in view of long-term sustainability. Furthermore, present N leaching data were only available for approximately 120 plots for which input output budgets could be calculated.

*Ground vegetation:* In the literature, the low acceptable N leaching rate is often taken to calculate critical N loads in view of possible vegetation changes (e.g. Downing et al., 1993), since literature data indicate that vegetation changes may take place in a situation where N leaching increases above natural background values (Van Dam, 1990). In reality, there is no direct relationship between N leaching and vegetation changes. It is the increase in N availability through enhanced N cycling that triggers the changes (e.g. Berendse et al., 1987). In this study, the use of natural N leaching rates did lead to calculated critical N deposition levels that are generally lower than empirical data on vegetation changes in forest (Section 4.2.2.1; Table 4.1. Consequently, we used a higher N concentration in view of vegetation changes (0.2 mol<sub>c</sub>.m<sup>-3</sup> or 2.8 g.m<sup>-3</sup>), but this value

was an approximate guess and more research is needed to come up with better criteria (see also Section 4.4).

*Trees:* Critical limits in view of impacts on trees have been derived in relation to nutrient imbalances. Examples are a critical  $NH_4/K$  ratio of 5 mol.mol<sup>-1</sup> in the soil solution, based on a strong decrease in the uptake of Ca and Mg above this value in a laboratory experiment with two-year-old Corsican pines, Boxman et al. (1988) or a critical N/K ratio in foliage of 4 g.g<sup>-1</sup> (11 mol.mol<sup>-1</sup>) related to K deficiency symptoms (Van den Burg,1988). For most coniferous tree species, an N concentration in the needles of 16 to 20 g.kg<sup>-1</sup> is considered optimal for growth. At these levels the sensitivity to frost and fungal diseases, however, increases, too. In a fertilisation experiment in Sweden, it was found that frost damage to the needles of Scots pine strongly increased above an N concentration of 18 g.kg<sup>-1</sup> (Aronsson, 1980). At this N level, the occurrence of fungal diseases such as *Sphaeropsis sapinea* and *Brunchorstia pinea* also appears to increase. In this study view of adverse impacts on coniferous trees, a critical limit of 18 g.kg<sup>-1</sup> in the needles was used to calculate critical loads. This critical N concentration was related to a critical dissolved N concentration in soil solution to allow the calculation of a critical leaching rate (see Section 4.2.4.2).

#### 4.2.3.2 Calculation methods

#### Calculation of long-term critical loads of nitrogen

Critical loads of total N for terrestrial ecosystems can be derived with a simple model of the N balance. A mass balance including all nitrogen fluxes (in mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>), neglecting the internal N turnover by litterfall and root uptake, reads (UNECE, 1996):

$$N_{td} = N_{gu} + N_{im} + N_{de} + N_{vo} + N_{er} + N_{ad} + N_{le} - N_{fi}$$
(4.1)

where the subscript *td* refers to total deposition, *fi* to fixation, *gu* to growth uptake, *im* to net immobilisation (further denoted as accumulation), *de* to denitrification, *vo* to volatilisation, *er* to erosion, *ad* to adsorption and *le* to leaching. Adsorption of N can, however, be neglected as this process only plays a temporary role. Furthermore, even in the short term, adsorption of N can generally be neglected since (i) N mainly occurs as  $NO_3^-$  except for the topsoil and (ii) the preference of the adsorption complex for  $NH_4^+$  is mostly low, especially in (acid) sandy soils. Volatilisation of N can play a role in grazed woodlands and in areas with frequent forest fires, whereas N removal by erosion may play a role at steep slopes. However, in most cases these N fluxes are negligible. N fixation is also small in most forest and heathland ecosystems except for N-fixing species, such as red alder (Van Miegroet and Cole, 1984). By assuming N fixation and N loss by volatilisation and erosion and N adsorption to be negligible, Eq. 4.1 can be written as (cf. De Vries, 1993):

$$N_{td} = N_{gu} + N_{im} + N_{de} + N_{le}$$
(4.2)

When defining the critical load via Eq. 4.2 it is implicitly assumed that all terms on the right-hand side do not depend on the deposition of nitrogen. This is probably not the case and thus all quantities should be taken "at critical load". However, to compute "denitrification at critical load" one needs to know the critical load, the very quantity one tries to compute. The only way to avoid this circular reasoning is to establish a functional relationship between deposition and the sink of 86

N, insert this function into Eq. 4.2 and solve for the deposition (to obtain the critical load). This has been done for denitrification. In the simplest case it is assumed to be linearly related to the net input of N by (De Vries, 1993; UNECE, 1996):

$$N_{de} = f_{de} \cdot (N_{td} - N_{gu} - N_{im})$$
(4.3)

where  $f_{de}$  ( $0 \le f_{de} \le 1$ ) is the so-called denitrification fraction, which has been formulated as a function of soil type (De Vries et al., 1994a). This equation is based on the assumption that the excess N input leaches as NO<sub>3</sub>. In most (forest) soils, N below the root zone (at 1m depth) is indeed dominated by NO<sub>3</sub>, even in the Netherlands with extremely high NH<sub>4</sub><sup>+</sup> inputs (De Vries, 1994) and therefore it is reasonable to assume that NH<sub>4</sub><sup>+</sup> leaching is negligible. This formulation implicitly assumes that accumulation and growth uptake are faster processes than denitrification. From Eq. 4.2 and Eq. 4.3 a critical N load, CL(N), can be derived:

$$CL(N) = N_{gu,crit} + N_{im,crit} + N_{le,crit}/(1 - f_{de})$$

$$(4.4)$$

where  $N_{gu,crit}$  stands for the average net uptake during the rotation period at the critical N load,  $N_{im,crit}$  stands for a critical (long-term acceptable) N accumulation and  $N_{le,crit}$  for a critical level of N leaching. The denitrification at critical load,  $N_{de,crit}$ , is implicitly related to the critical N leaching according to Eq. (4.4). In wet forest and heathland soils, deep ground water and surface waters denitrification is generally not negligible and should be accounted for.

In words, Eq. (4.4) states that the critical load of nitrogen equals a critical N leaching rate from the soil plus a critical N retention, where N retention stands for the sum of uptake, accumulation and denitrification. It is based on the assumption that an N input above a net N uptake by forest growth, long-term acceptable N accumulation, denitrification and an acceptable rate of N leaching causes adverse impacts. This will either lead to N accumulation in the system (violation of the stand-still principle) or to unacceptable high N contents in foliage, soil and soil solution, thus causing adverse affects such as nutrient imbalances, increased sensitivity to frost, drought and diseases and vegetation changes (effect-based principle).

# Calculation of the critical nitrogen leaching rate

Crucial in the calculation of the critical or acceptable N leaching rate. In this study, this rate is calculated by multiplying the precipitation excess with a critical N concentration according to:

$$N_{le(crit)} = 10000 \cdot PE \cdot [N]_{ss(crit)}$$

where:

PE = the precipitation excess, being the flux of water leaching from the root zone  $(m.yr^{-1})$ [N]<sub>ss(crit)</sub> = the critical limit for the total concentration of nitrogen in the percolating soil solution

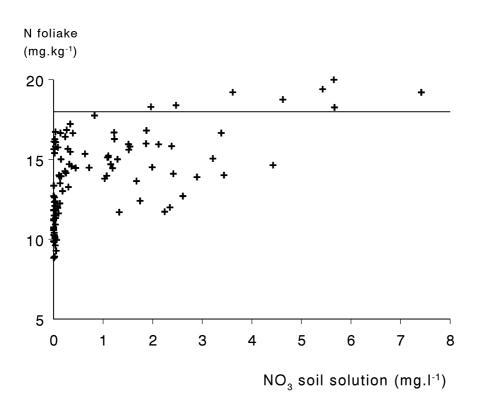
 $_{it}$  = the critical limit for the total concentration of nitrogen in the perc (mol<sub>c</sub>.m<sup>-3</sup>)

The value of 10000 is needed to convert the unit from  $mol_c.m^{-2}.yr^{-1}$  to  $mol_c.ha^{-1}.yr^{-1}$ . In the standstill approach the present N leaching loss for the plots with input-output budgets was used. Using the current N leaching implies that critical loads can become high on systems that are N saturated (here defined as systems that do not accumulate N any more) due to decades of high N inputs. This is an undesirable situation in view of ecosystem protection and long-term sustainability.

(4.5)

Therefore, an alternative calculation was also carried out, using a value for  $N_{ss(crit)}$  of 0.02 mol<sub>c</sub>.m<sup>-3</sup>, based on the runoff under pristine conditions. Actually, the use of a natural N leaching rate is not really related to a stand-still approach, but it gives information about the accumulation that either now or in the past (has) taken place.

Effect-based critical loads were calculated by using a value of  $0.2 \text{ mol}_{c}.m^{-3}$  in view of possible impacts on ground vegetation (Section 4.2.3.1). Actually, a value of  $0.02 \text{ mol}_{c}.m^{-3}$  has also been used in the literature, but critical loads thus calculated are too low compared to empirical critical loads for vegetation changes (see Section 4.3). The critical dissolved N concentration in view of adverse impacts on forests was related to a critical N concentration in the needles of 18 g.kg<sup>-1</sup>, above which the sensitivity to frost and fungal diseases increases (Section 2.2). In this study, we derived the relationship on the basis of the results for the considered Intensive Monitoring plots, as shown in figure 4.5. From this graph the critical limit in the soil solution was estimated as 3.3 mg.l<sup>-1</sup> being the value above which most N contents in foliage exceed 18 g.kg<sup>-1</sup>.



*Figure 4.5 Relationship between measured N content in the foliage of coniferous trees and the average NO*<sub>3</sub> *concentration in soil solution for approximately 120 Intensive monitoring plots.* 

#### Adaptations to calculate present deposition thresholds

The major premise of the calculation of a critical load described above is that it assumes a steadystate situation to derive a long-term acceptable load. Consequently, the different N fluxes are calculated for a long-term average situation, such as (De Vries, 1993; UNECE, 1996):

- The rotation period for the calculation of N uptake
- An acceptable accumulation rate of stable organic N compounds in the soil (stable forms of humus) in e.g. the period since glaciation for the calculation of N immobilisation
- A 30-year average precipitation excess for the calculation of the critical N leaching rate

To gain insight in the relationship in e.g. the crown condition and atmospheric deposition, however, we need to know the actual threshold at the time of consideration. This implies that the magnitude of processes influencing nitrogen retention and release at present have to be estimated. An actual assessment can be based on the results of input-output budgets, which do give actual information on the retention of nitrogen in a forest ecosystem. Such budgets have been made for 121 Intensive monitoring plots for the period 1995-1998 (De Vries et al., 2001).

The differences in the calculation of critical loads and present deposition thresholds for N are listed in Table 4.4. The calculation of the long-term critical loads was based on the approach described above, with the exception that we did not use a 30-years average value for the precipitation excess. Instead, calculated values for the period 1995-1998 were used, because data needed to compute site specific leaching fluxes for longer periods were not available. Instead of using a long-term average value for the calculation of uptake, accumulation and denitrification separately, we considered the sum of these processes in the period 1995-1998 in the assessment of a present deposition threshold. This sum, being denoted as the total N retention, was derived from the difference between the N input by deposition and the N output by leaching.

Considered process	Steady state long-term critical loads <sup>1)</sup>	Present deposition thresholds <sup>2)</sup>
Nitrogen retention	Calculated as uptake plus accumulation plus denitrification as specified below in this table.	Estimated by subtracting N leaching (based on measured N concentrations and calculated precipitation excesses) from the measured N deposition (based on throughfall and stemflow) in the period 1995-1998 and scaling the result <sup>3</sup>
Uptake of nitrogen	Multiplication of average yield during the rotation period with literature values for the N contents in stemwood	_ 4
Nitrogen immobilisation/ accumulation	Long-term acceptable value according to critical load Mapping Manual (UNECE, 1996)	_4
Denitrification	Related linearly to net N input at critical load (in this approach to the critical N leaching)	_4
Critical N leaching rate	Multiplication of calculated actual precipitation excess in period 1995-1998 <sup>5</sup> with a critical N concentration.	Multiplication of calculated actual precipitation excess in period 1995-1998 <sup>5</sup> with a critical N concentration.

 Table 4.4
 Difference in the calculation of long-term critical loads and present deposition thresholds for nitrogen

1) Steady state long-term critical loads were calculated for all plots for which deposition data and hydrological fluxes were available (234 plots; see Section 4.2.1)

2) Present deposition thresholds were calculated for plots for which both deposition and soil solution chemistry were available (starting with 121 plots with data on element budgets, calculations could ultimately be made for .. plots

3) The present N retention was scaled to the N retention that would occur at an N input that is equal to the present deposition threshold.

4) The separate N fluxes in terms of uptake, accumulation and denitrification could not be derived from the measurements. Instead, we could only derive the sum of these N fluxes, in terms of total N retention, from the difference between N deposition and N leaching.

5) A long-term (e.g. 30-years average) value for the precipitation excess would have been favoured but data were only available for a four-year period of 1995-1998.

Actually, in calculating the present deposition threshold, one has to know the N retention that would occur at an N input that is equal to the present deposition threshold,  $N_{ret,crit}$ , according to:

$$PDT(N) = N_{ret,crit} + N_{le,crit}$$

(4.6)

If one would use the present N retention, which takes place at the present N input, one would assume that the N retention is independent of the N input, which is an invalid assumption. Consequently, the present N retention has to be scaled to the N retention at the present deposition threshold.

We investigated two possibilities to calculate the present deposition threshold. The first was to scale the N retention by deriving an empirical relationship between N retention and N deposition, in combination with other environmental characteristics, such as soil type and soil C/N ratios, according to (see previous Technical Report: De Vries et al., 2001):

$$N_{ret} = \alpha_0 + \alpha_1 x_1 + \dots + \alpha_n N_{dep}$$
(4.7)

where  $N_{ret}$  is the expectation value of the N retention, being the response variable,  $x_1$  to  $x_{n-1}$  are predictor variables (tree species, soil type, humus type, climatic region, altitude, stand age, the fraction of ammonium in the deposition, pH and C/N ratios in both the organic layer and the mineral topsoil) and  $\alpha_1$  to  $\alpha_{n-1}$  are the regression coefficients. The regression relations were carried out by using both the original data and after a logarithmic transformation of both the N retention and N deposition data, since those data that were highly skewed. Using the original data, the N retention at a given site in dependence of the N deposition was calculated according to:

$$N_{ret} = A + B \cdot N_{dep} \text{ or } N_{ret,crit} = A + B \cdot PDT(N)$$
(4.8)

In this formula, A is a site-specific value, that is calculated by applying Eq. (4.8), inserting the site specific data for tree species, soil type, humus type etc, multiplied by the ratio of the measured and predicted N retention. B is regression coefficient  $\alpha_n$  multiplied by the ratio of the measured and predicted N retention. Inserting Eq. (4.8) in Eq (4.6) and gives:

$$PDT(N) = A + N_{le,crit} / (1 - B)$$

$$(4.9)$$

We also investigated the possibility to calculate the present deposition threshold by directly relating the N leaching to the N deposition according to:

$$N_{le} = \alpha_0 + \alpha_1 x_1 + \dots + \alpha_n N_{dep}$$

$$(4.10)$$

where  $N_{le}$  is the expectation value of the N leaching, and all other variables being equal to those mentioned under Eq. (4.7). As with N retention, the regression relations were carried out by using both the original data and after a logarithmic transformation of both the N leaching and N deposition data. As with N retention, N leaching at a given site in dependence of the N deposition was calculated according to:

$$N_{le} = C + D \cdot N_{dep} \text{ or } N_{le,crit} = C + D \cdot PDT(N)$$
(4.11)

Similar to the N retention, C is a site-specific value, that is calculated by applying Eq. (4.10) and multiplying the result by the ratio of the measured and predicted N leaching, whereas D is regression coefficient  $\alpha_n$  multiplied by the ratio of the measured and predicted N leaching. Use of Eq. (4.11) directly leads to an estimate of PDT(N) according to:

$$PDT(N) = (N_{le,crit} - C)/D$$

$$(4.12)$$

By using the direct empirical approach between N leaching and N deposition, the N retention at present deposition threshold is indirectly accounted for. In both Eq (4.9) and Eq (4.12), the value of  $N_{le,crit}$  was derived by multiplying the calculated precipitation excess with a critical N concentration.

### 4.2.3.3 Assessment of input data

Below we first describe the assessment of input data for the calculation of long term critical nitrogen loads and then the approach used for the assessment of present deposition thresholds.

#### Nitrogen uptake

In assessing the critical load, nitrogen uptake is the net growth uptake, i.e. the net uptake by vegetation that is needed for long-term average growth. Nitrogen input by litterfall and nitrogen removal by maintenance uptake (needed to re-supply nitrogen to leaves) is not considered here, assuming that both fluxes are equal in a steady-state situation. Thus the net uptake is calculated, being equal to the annual average removal in harvested biomass. In this calculation we assumed that this includes the removal of stems only (no branches or leaves).

Net nitrogen uptake was calculated at each site by multiplying the annual average growth rate of stems with the stem density and the nitrogen content in stems. For the densities of stemwood and the nitrogen contents in stems use was made of literature data (e.g. Kimmins et al., 1985 and De Vries et al., 1994a,b), as presented in Table 4.5. This table also includes the BC contents that were used in the calculation of base cation uptake that is needed to calculate critical acid loads.

Tree species	Stem density (kg.m <sup>-3</sup> )	N content in stemwood (g.kg <sup>-1</sup> )	BC content in stemwood (g.kg <sup>-1</sup> )
Scots Pine	510	1.2	1.6
Norway Spruce	460	1.2	1.2
Oak	700	1.7	2.5
Beech	700	1.4	2.4

 Table 4.5
 Average values used for the densities of stemwood and the N and BC (Ca+Mg+K) contents in stemwood of the considered main tree species

The annual average growth rate of stems (the yield) was base on site specific data yield data, given in intervals of 5  $m^3$ .ha<sup>-1</sup>.yr<sup>-1</sup>. In future calculations, we might improve this preliminary estimate by taking the ratio of the present standing volume, derived by data on diameter at breast height and tree height, and the stand age of the tree. This approach makes optimal use of the site data, but it does not gives the average yield for the whole rotation period, but the average yield since the time of germination.

# Nitrogen immobilisation

As with uptake,  $N_{im}$  denotes the long-term net immobilisation (accumulation) of nitrogen in the soil, i.e. only the continuous build-up of stable C/N-compounds in forest soils. Using data from Swedish forest soils, Rosén et al. (1992) estimated the annual nitrogen immobilisation since the last glaciation at 0.2-0.5 kg N.ha<sup>-1</sup>.yr<sup>-1</sup>. Values between 0.5 and 1.0 kg N.ha<sup>-1</sup>.yr<sup>-1</sup> are currently recommended for the critical loads work under the LRTAP Convention (UNECE, 1996).

### **Denitrification**

Denitrification was described as a fraction of the net input to the soil minus the calculated net immobilisation (see Eq. 4.3). Denitrification fractions were related to soil clusters based on data given in Breeuwsma (1991) for agricultural sand, clay and peat soils in The Netherlands. Data were corrected for the more acid circumstances in forest soils. Values thus used are 0.8 for peat soils, 0.7 for clay soils (texture classes 2, 3 and 2/3), 0.5 for sandy soils (texture classes 1 and 1/2) with glevic features and 0.1 for sandy soils without glevic features.

### Critical N leaching

The natural N leaching rate was calculated by multiplying the precipitation excess by a critical dissolved N concentration as described in Section 4.2.3.2. The precipitation excess was calculated with a water balance model, as described in De Vries et al. (2001). The critical dissolved N concentration was set equal to (section 4.2.3.1):

- A background value of 0.02 mol<sub>c</sub>.m<sup>-3</sup>, in view of N accumulation in forest soils.
- An estimated value of 0.20 mol<sub>c</sub>.m<sup>-3</sup> in view of impacts on ground vegetation. An estimated value of 0.24 mol<sub>c</sub>.m<sup>-3</sup> in coniferous forests, related to a critical N concentration in the needles of 18 g.kg<sup>-1</sup>, above which the sensitivity to frost and fungal diseases increases.

### **Present deposition thresholds**

After investigating the various possibilities to derive a present deposition threshold, described in Section 4.2.4.2, we based the value on a direct relationship between N leaching and N deposition, using the original data (no logarithmic transformation) according to (compare Eq. 4.10):

$$N_{le} = -68.6 + 103 \cdot fNH_4 + 0.2346N_{dep}$$
(4.13)

where  $fNH_4$  is the fraction of ammonium in the deposition. In the ultimate equation used (4.13) this fraction appeared to be the most influential factor in the assessment of the N leaching rate as a function of N deposition. The value of PDT(N) was calculated with Eq (4.12) according to the procedure described in that section.

# 4.2.4 Calculation of critical loads and present deposition thresholds for acidity

In this section, first an overview is presented of the impacts of acidity on forest ecosystems, followed by the critical limits thus derived and used in this study (Section 4.2.4.1). The background of the derivation of the simple mass balance model for acidity, used to calculate critical acid loads in this study, is presented in Section 4.2.4.2. This section also includes methods to calculate of the critical acidity leaching rate and the present deposition threshold. The input data for the calculations are described in Section 4.2.4.3.

# 4.2.4.1 Impacts of acidity and critical limits

# Impacts on forest ecosystems

The indirect soil-mediated acidifying impacts of S and N deposition include the loss of base cations from the soil and the release of soluble toxic aluminium. In the 1980s, several authors (e.g. Ulrich et al., 1979, Hutchinson et al., 1986) considered soil acidification responsible for forest decline, since Al<sup>3+</sup> is toxic to plant roots (Cronan et al., 1989; Marschner, 1990; Mengel, 1991; Cronan and Grigal, 1995). This direct relationship has been questioned, since there is no clear relationship between forest crown condition and high Al concentrations or Al/base cation ratios (e.g. Hendriks et al., 1997). Nevertheless, numerous studies, both in the laboratory and in the field, have shown that high concentrations of Al relative to (divalent) base cations such as Ca and Mg, have a negative influence on mycorrhizal frequency, root elongation and root uptake (see Sverdrup and Warfvinge, 1993, for an overview). It may affect fine root growth, thus inhibiting the uptake water and of base cations, causing deficiencies of these nutrients for forest trees (notably Mg).

Roelofs et al. (1985) and Schulze (1989) suggested that acidification of soil and excessive N inputs caused nutrient imbalances in the soil and the plants. This coincided with field observations and foliar analyses, indicating that deficiencies of Mg and K cause yellowing of needles of Norway spruce (Bosch et al., 1983; Zöttl and Mies, 1983). Roberts et al. (1989) concluded that spruce decline in Central Europe mainly results from foliar Mg deficiency due to (i) an increased Mg demand induced by an increased growth in response to elevated N inputs, and (ii) inhibition of Mg uptake caused by soil acidification (a decrease in exchangeable magnesium, ammonium accumulation and aluminium mobilisation). In general, N contributes both to soil acidification and eutrophication and the two processes lead to the imbalance in nutrient availability for plant growth (e.g. Heij and Erisman, 1997).

Apart from impacts on forest, acid deposition may affect the species diversity of terrestrial vegetation, due to a decrease in pH (Van Hinsberg and Kros, 1999), cause pollution of groundwater for drinking water supply due to increased hardness and increased concentrations of Al mobilised from the soil (e.g. Boumans and Van Grinsven, 1991) and lead to loss of fish populations caused by a decrease in pH and an increase in labile Al in surface waters (e.g. Hultberg, 1988).

# Critical limits for forest ecosystems

The most common critical chemical value used in the European critical loads work is the molar ratio of base cations to aluminium in soil solution in view of impacts on tree roots and thereby on forest condition. Sverdrup and Warfvinge (1993) derived critical limits for Al/(Ca+Mg+K) ratios for many vegetation types, based on a literature review of numerous laboratory studies, relating those ratios to decreased root biomass and inhibited nutrient uptake. Critical limits for Al/(Ca+Mg+K) ratios were based on that review and equalled 0.8 for pine and spruce and 1.6 for oak and beech (see also Table 4.3) indicating that the conifers are more sensitive to aluminium toxicity that the broadleaves. Although the critical base cation to aluminium ratio is the most widely used critical chemical value in the ongoing critical loads work, it is not undisputed. More information on this point is given in Section 6.2.2. At sites where low Bc concentrations, where the use of a critical Al/Bc ratio would lead to very low (non-toxic) Al concentrations, we directly used a critical aluminium concentration, [Al]<sub>crit</sub> of 0.2 mol<sub>c</sub>.m<sup>-3</sup> (after Cronan and Grigal, 1995).

#### 4.2.4.2 Calculation methods

#### Calculation of long-term critical loads of acidity

In this report acidity was simply defined as the sum of sulphate and nitrogen. In the literature, acidity have also been defined by subtracting the sea salt corrected base cation input from the sum of sulphate and nitrogen, implying that that this also has to be done for the critical load (e.g. De Vries et al., 1994a). Furthermore, some countries only subtract sea salt corrected base cation bulk deposition, which means that one has to correct for dry deposition input in the critical load calculation (e.g. De Vries et al., 1994b). It is considered most simple to lump S and N on one hand and add alkalinity in the atmospheric deposition in the CLO calculation, and consequently this approach was used in this report. Critical loads of acidity, induced by deposition of N and S, can be derived from the steady-state charge balance for the ions in the soil leachate (in  $mol_c.ha^{-1}.yr^{-1}$ ) leaving the root zone (modelled as a single homogeneous layer):

$$H_{le} + Al_{le} + BC_{le} + NH_{4,le} = SO_{4,le} + NO_{3,le} + Cl_{le} + HCO_{3,le} + RCOO_{le}$$
(4.14)

where  $\text{RCOO}_{\text{le}}$  is the leaching flux of the sum of organic anions. Neglecting  $\text{OH}^-$  and  $\text{CO}_3^{2-}$  (a reasonable assumption even for calcareous soils), the alkalinity or ANC (Acid Neutralising Capacity) can be defined as:

$$ANC_{le} = HCO_{3,le} + RCOO_{le} - H_{le} - Al_{le}$$

$$(4.15)$$

A steady-state situation with respect to acidification implies a constant pool of exchangeable base cations (BC). Consequently the following mass balance holds for base cations:

$$BC_{le} = BC_{td} + BC_{we} - Bc_{gu}$$
(4.16)

Note, that  $BC_{td}$  and  $BC_{we}$  include all four base cations (BC=Ca+Mg+K+Na), whereas sodium is not taken up by vegetation (Bc=BC-Na). For sulphur the mass balance reads:

$$S_{le} = S_{td} - S_{gu} - S_{im} - S_{re} - S_{ad}$$
(4.17)

where the subscript re refers to reduction. An overview of S cycling in forests by Johnson (1984) suggests that the net (growth) uptake, immobilisation, and reduction of  $SO_4^{2^-}$  are generally insignificant. Sulphate adsorption occurs especially in Fe- and Al-oxide rich subsurface horizons (Johnson et al., 1979, 1982; Johnson and Todd, 1983). However, when deriving a long term critical load, the effect of adsorption must be neglected, since this phenomenon is only of temporary importance (several decades). Even in the short term, sulphate adsorption is negligible in most European forest ecosystems, as shown by various budget studies given in Berdén et al. (1987). Furthermore, since sulphur is completely oxidised in the soil profile, S<sub>le</sub> equals  $SO_{4,le}$ , and Eq. 4.9 simplifies to:

$$SO_{4,le} = S_{td} \tag{4.18}$$

Combining Eqs. 4.7, 4.8 and Eq. 4.10 and the N balance derived in section 4.3 (Eq. 4.2;  $NO_{3,le}=N_{le}$ ) yields for the charge balance (Eq. 4.5):

$$S_{td}^{*} + N_{td} = BC_{td}^{*} + BC_{we} - Bc_{gu} + N_{gu} + N_{im} + N_{de} - ANC_{le}$$
(4.19)

where the star denotes Cl-corrected quantities, assuming that chloride comes only from sea spray. It is assumed that there are no sources or sinks of chloride in the soil compartment, and therefore leaching equals deposition. Knowledge of the deposition terms, weathering and net uptake of base cations as well as nitrogen uptake, accumulation and denitrification allows to calculate the ANC leaching, and thus assess the acidification status of the soil. Conversely, critical loads of S and N can be computed by defining a critical (or acceptable) ANC leaching, ANC<sub>le,crit</sub>, which is set to avoid "harmful effects" on a "sensitive element of the environment" (e.g. damage to fine roots).

$$CL(S+N) = BC_{td}^{*} + BC_{we} - Bc_{gu} + N_{gu} + N_{im} + N_{de} - ANC_{le,crit}$$
 (4.20)

In words, Eq. (4.11) states that the critical acid load equals a critical acidity leaching rate from the soil plus the buffering of acidity by net base cation input (deposition plus weathering minus uptake) and N retention (uptake plus accumulation plus denitrification) at this critical leaching rate. Crucial in the calculation is the choice of the critical limit determining the critical acidity leaching rate. Differences in critical load values for forest soils, groundwater and surface waters in the same area are mainly due to differences in the weathering rate, denitrification rate and the critical alkalinity leaching flux. The areal weathering rate, expressed in mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>, is determined by the parent material and the considered depth of the soil profile.

#### Derivation of separate critical loads for sulphur and nitrogen related to soil acidification

Using the equation for the deposition-dependent denitrification (Eq. 4.3), one obtains for the critical loads of sulphur, CL(S), and acidifying nitrogen, CL(N):

$$CL(S) + (1 - f_{de}) \cdot CL(N) = BC_{td}^{*} + BC_{we} - Bc_{gu} + (1 - f_{de}) \cdot (N_{im} + N_{gu}) - ANC_{le,crit}$$
(4.21)

Note, that these critical loads of S and N are not unique; every pair of deposition ( $N_{td}$ ,  $S_{td}$ ) which fulfils Eq. 4.12 are critical loads. However, when comparing S and N deposition to critical loads one has to bear in mind that the nitrogen sinks cannot compensate incoming sulphur acidity, i.e. the maximum critical load of sulphur is given by:

$$CL_{max}(S) = BC_{td}^* + BC_{we} - Bc_{gu} - ANC_{le,crit}$$
(4.22)

Furthermore, if

$$N_{td} \le N_{im} + N_{gu} = CL_{min}(N)$$
(4.23)

all deposited N is consumed by uptake and immobilisation, and sulphur can be considered alone. The maximum amount of allowable N deposition (in case of zero S deposition) is given by (see Eqs. 12-14):

$$CL_{max}(N) = CL_{min}(N) + CL_{max}(S)/(1 - f_{de})$$
 (4.24)

#### Calculation of the critical ANC leaching when using a critical Al/(Ca+Mg+K) ratio

Selecting a critical chemical value is the most crucial step in calculating critical loads, since this quantity links deposition level to a "harmful effect". For acidity, the critical leaching of ANC has to be specified. Criteria for [Al] generally refer to the inorganic Al concentration, which is toxic to roots. Assuming that organic anions are completely bound by Al, Eq. 4.6 simplifies to:

$$ANC_{le,crit} = Q \cdot ([HCO_3] - [Al]_{crit} - [H]_{crit})$$
(4.25)

Where Q is the percolation flux  $(m.yr^{-1})$ . The concentration of HCO<sub>3</sub> can be derived by an equilibrium equation describing the dissociation of CO<sub>2</sub> according to:

$$[\text{HCO}_3] \cdot [\text{H}] = \text{K}_{\text{CO2}} \cdot \text{p}_{\text{CO2}} \tag{4.26}$$

where  $K_{CO2}$  is the dissociation constant and  $p_{CO2}$  the partial pressure of  $CO_2$ . The concentration of bicarbonate can be neglected in calculating critical loads for acid forest soils, where the critical pH is below 5.

The acceptable concentration of Al can be derived by relating it to the base cation leaching. The critical Al/Bc ratio (where Bc=Ca+Mg+K) is here used to calculate a critical aluminium leaching according to:

$$A_{le, crit} = 1.5 \frac{(Al/Bc)_{crit}}{Bc_{le}} \quad \text{with} \quad Bc_{le} = Bc_{td} + Bc_{we} - Bc_{gu}$$
(4.27)

where  $BC_{le}$  is the base cation (Ca+Mg+K) leaching rate in mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. The factor 1.5 arises from the conversion of the molar Al/ Bc ratio to equivalents. Note that weathering generally includes Na, but this is taken care of by multiplying the base cation weathering rate by a factor of 0.85. Dividing by Q, the critical Al concentration is obtained. The same procedure can be used to compute Al<sub>le,crit</sub> from a critical Al/ Ca or Al/(Ca+Mg) ratio. The values used in this study for Al/ Bc<sub>crit</sub> are 0.8 for conifers and 1.6 for broadleaves, respectively (see Table 4.3).

Finally, the H concentration can be calculated form the Al concentration, using an equilibrium equation describing the dissolution of aluminium (hydr)oxides, according to:

$$[AI] = KAI_{ox} \cdot [H]^a \tag{4.28a}$$

or inversely:

$$[H] = ([AI]/KAI_{ox})^{1/a}$$
(4.28b)

where  $\alpha$  is a site-dependent exponent indicating the type of interaction between Al and protons on binding sites in the soil. Values between 1 and 2 do indicate interactions between Al and protons on soil organic matter. For  $\alpha$ =3 this is the familiar gibbsite equilibrium, and KAl<sub>ox</sub>=K<sub>gibb</sub>, being the Gibbsite equilibrium constant, that is mostly used to calculate critical loads. Values for logK<sub>gibb</sub> generally vary between 8.0 and 8.5.

Use of the gibbsite equilibrium for modelling Al concentrations seems to work well enough for the B horizon of podzols (although equilibrium with imogolite has also been suggested), but it doesn't work well for organic horizons. At pH values below 5, organic horizons may be undersaturated with respect to gibbsite and Al solubility is controlled by organic complexation (De Wit, 2000). Complexation reactions with soil organic matter may even control the solubility of Al in the upper part of some B horizons as well (Simonsson, 1999). A further problem with the use of the gibbsite equilibrium model arises from the fact that Al(OH)<sub>3</sub> occurs in many different forms, of which the one most commonly occurring in Nordic soils is amorphous, and certainly not a well-defined crystalline phase. Its solubility constant is not the same as that of crystalline gibbsite (Simonsson, 1999).

Considering these aspects, it is more accurate to use values for  $\alpha$  that are lower than 3, and consequently also a deviating Kal<sub>ox</sub> value, which refers to an amorphous Al hydroxide and not to Gibbsite. Regression analyses between the negative logarithm of the Al concentration (pAl) against the negative logarithm of the H concentration (pH), with concentrations in mol.l<sup>-1</sup> for nearly 300 Dutch forest stands give results as presented in Table 4. (Van der Salm and De Vries, 2001). The data are based on measurements in approximately 200 sandy soils (Leeters and de Vries, 2001) and in approximately 40 loess soils, 30 clay soils and 30 and peat soils (Klap et al., 1999). The results for the subsoil show that the standard gibbsite equilibrium constant and exponent of 3.0 is reasonable for clay soils. Very different values, however, have to be used for peat soils and to a lesser extent also for sandy and loess soils. This is especially true at lower soil depths (not given in Table 4.6) but the model was only applied by calculating the leaching rate from the bottom of the rootzone.

Table 4.6	Estimated values of KAlox and $\alpha$ for the subsoil (30-100)
	cm) based on regression between pAl and pH in the soil
	solution of Dutch forest soils

	solution of D	uich joresi s	ons		
Soil type	$\log KAl_{ox}$	α	R <sup>2</sup> <sub>adj</sub>	n	
Sand	5.20	2.51	86.3	172	
Loess	4.55	2.17	92.4	39	
Clay	7.88	2.65	87.3	116	
Peat	-1.06	1.31	81.2	116	

Thus, specifying a critical Al/Bc ratio, the critical ANC leaching can be computed by calculating the related Al and H concentration. This approach is questionable, however, at sites with very low Bc concentrations. There, a very low critical Al concentration is calculated as well, which is unlikely to be toxic. At those sites, we directly used a critical aluminium concentration,  $[Al]_{crit}$  of 0.2 mol<sub>c</sub>.m<sup>-3</sup> (see before). In the stand-still principle, the acceptable critical Al concentration has to be derived indirectly from the present base saturation or an acceptable Al weathering rate, as discussed below.

# Calculation of the critical ANC leaching from a critical base saturation

Base saturation, i.e. the fraction of base cations on the cation exchange complex, is an indicator of the acidity status of a soil and one may want to keep this pool to avoid nutrient deficiencies in the long term. In the following we show how to link base saturation with ANC, thus allowing the use of a critical base saturation or the present base saturation for critical load calculations based on the stand-still principle (sustainable base cation pools).

Cation exchange reactions are often described by the Gaines-Thomas equations. Neglecting Naexchange and lumping Ca, Mg and K, the following equations describe the Bc-Al-H exchange:

$$\frac{E_{Al}^{2}}{E_{Bc}^{3}} = K_{AlBc} \frac{[Al]^{2}}{[Bc]^{3}}$$
(4.29)

$$\frac{E_{\rm H}^2}{E_{\rm Bc}} = K_{\rm HBc} \frac{[{\rm H}]^2}{[{\rm Bc}]}$$
(4.30)

where  $E_X$  is the exchangeable fraction of ion X, and  $E_{Bc}$  in particular is the base saturation. Charge balance requires that

$$E_{Bc} + E_{Al} + E_{H} = 1$$
 (4.31)

A relationship between the base saturation and the Al/Bc-ratio can be easily derived (with [Bc] as additional parameter). Inserting  $E_{Al}$  and  $E_{H}$  from Eqs.(4.29) and (4.30) into Eq.(4.31) and using Eq.(4.28b) to express [H] in terms of [Al] yields:

$$E_{Bc} + E_{Bc}^{3/2} \frac{r_{eq} K_{AlBc}^{1/2}}{[Bc]^{1/2}} + E_{Bc}^{1/2} \frac{r_{eq}^{1/a} K_{HBc}^{1/2}}{KAl_{ox}^{1/a} [Bc]^{1/2-1/a}} = 1$$
(4.32)

where  $r_{eq}=[Al]/[Bc]$  stands for the equivalent Al/BC ratio. This third-order equation in  $E_{Bc}^{1/2}$  can be solved numerically to obtain the base saturation as function of the (critical) Al:Bc ratio and thus relate it to the (critical) ANC leaching.

### Calculation of the critical ANC leaching in view of sustainable aluminium pools

A critical ANC value can also be calculated by aiming at a negligible depletion of Al-hydroxides (Al depletion criterion), being a relevant precautionary approach in acid soils with a low base saturation. This approach is based on the idea that using an ANC limit based on a critical Al concentration or Al/Bc ratio may imply that the accepted rate of Al leaching is greater than the rate of Al mobilisation by weathering of primary minerals. The remaining part of Al has to be supplied from readily available Al pools including Al hydroxides. This causes depletion of these minerals, which might induce an increase in Fe buffering which in turn leads to a decrease in the availability of phosphate (De Vries, 1994). Furthermore, the decrease of those pools in podzolic sandy soils may cause a loss in the structure of those soils.

Negligible depletion of Al hydroxides is achieved when Al leaching equals mobilisation of Al from primary minerals. [Al]<sub>crit</sub> can thus be calculated as

$$[AI]_{crit} = r \cdot BC_{we} / Q \tag{4.33}$$

where r is the equivalent stoichiometric ratio of Al to BC in the congruent weathering of silicates (primary minerals). The other terms in the ANC (Eq. 4.16) can again be derived from the equilibrium equations discussed before (Eqs. 4.26 and 4.28b).

#### Adaptations to calculate present threshold depositions

As with nitrogen, the major premise of the calculation of a long-term critical acid load is that it assumes a steady-state situation. To gain insight in the present impacts of atmospheric deposition, we need to know the threshold above which effects might occur now. This implies that the magnitude of all processes buffering the incoming acidity at present have to be estimated. This includes all processes influencing base cation release, such as weathering and cation exchange, nitrogen retention and sulphur retention or release. In the assessment of a steady-state long-term critical load, dynamic processes such as cation exchange and sulphur retention or release have been neglected, whereas nitrogen retention refers to a steady-state situation and not to the present state. As stated before, an actual assessment can also be based on the results of input-output budgets for sulphur, nitrogen and base cations. Such budgets, that have been made for 121 Intensive monitoring plots for the period 1995-1998 (De Vries et al., 2001), do give actual information on the retention or release of those elements in a forest ecosystem.

The differences in the calculation of the critical loads and the present deposition thresholds of acidity are listed in Table 4.7. In the calculation of a steady-state critical load, we used long-term average values for the calculation of each model input. Instead, in deriving the present deposition threshold, we considered the time period 1995-1998 in deriving the base cation leaching, N retention/release and S retention/release, to account for the buffer processes at the time of consideration. These buffer processes were then scaled to the values that would occur at the present deposition threshold.

Considered process	Steady state long-term critical loads <sup>1)</sup>	Present threshold depositions <sup>2)</sup>
Base cation leaching	Calculated as deposition plus weathering	Actual average leaching in period 1995-
	minus uptake as specified below	1998 based on measured BC concentrations
		and calculated precipitation excess $^{3)}$
Total base cation deposition	Based on measured throughfall/ stemflow in	_3)
	period 1995-1998	_3)
Base cation weathering	Related to soil type, corrected for measured	
	temperatures in period 1995-1998 or interpolated values for a 10 year period	
Uptake of base cations	Multiplication of average yield during	_3)
optake of base cations	rotation period with literature values of BC	-
	contents	
Nitrogen retention/release	Calculated as uptake plus immobilisation	Actual average N deposition (based on
	plus denitrification as specified in Table 2	throughfall and stemflow) minus N leaching
		(based on measured N concentrations and
		calculated precipitation excess) in period
		1995-1998 <sup>4</sup>
Sulphate retention/release	Not included	Actual average $SO_4$ deposition (based on
		through fall and stem flow) minus $SO_4$
		leaching (based on measured SO <sub>4</sub>
		concentrations and calculated precipitation excess) in period 1995-1998 <sup>4</sup>
Critical Al leaching rate	Multiplication of calculated Bc leaching rate	Multiplication of (scaled) measured Bc
Critical Ai leaching fate	with a critical Al/Bc ratio	leaching rate with a critical Al/Bc ratio
1) Steady state long term cri	tical loads were calculated for all plots for which	

Table 4.7Difference in the calculation of long-term critical loads and present deposition thresholds for acidity

<sup>1)</sup> Steady state long-term critical loads were calculated for all plots for which deposition data and hydrological fluxes were available (226 plots; see Section 4.2.1)

<sup>2)</sup> Present deposition thresholds were calculated for 84 plots for which both deposition and soil solution chemistry were available and the pH was less than 4.5.

<sup>3)</sup> The separate BC fluxes in terms of deposition, weathering and uptake are not needed in this calculation, in which present or scaled Bc leaching fluxes are used.

4) The sum of the present N and S retention or release has to be scaled to the retention that would occur at an acid (S+N) input that is equal to the present deposition threshold

Comparable to nitrogen, in calculating the deposition threshold, one has to know the acid buffering (the sum of N and S retention or release) that would occur at an acid input that is equal to the present deposition threshold,  $(S+N)_{ret,crit}$ , according to (compare Eq. 4.20):

$$PDT(S+N) = BC_{le} + (S+N)_{ret,crit} - ANC_{le,crit}$$

$$(4.34)$$

As with N retention, this approach implies that the acid buffering which takes place at the present acid input, has to be scaled to the acid buffering at the present deposition threshold.

Unlike nitrogen, we did not investigate the possibility to calculate the present deposition threshold by deriving an empirical relationship between acid buffering and acid deposition, since the combination of S and N retention or release would lead to complex relationships and consequently a complex assessment of the PDT value. Instead, a direct relationship between Al leaching and acid (S+N) deposition was derived according to:

$$Al_{le} = \alpha_0 + \alpha_1 x_1 + \dots + \alpha_{n-1} x_{n-1} + \alpha_n (S + N)_{dep}$$
(4.35)

where  $Al_{le}$  is the expectation value of the Al leaching,  $x_1$  to  $x_{n-1}$  are predictor variables (tree species, soil type, humus type, climatic region, stand age, the fraction of ammonium in the deposition, pH and base saturation in the subsoil) and  $\alpha_1$  to  $\alpha_{n-1}$  are the regression coefficients (see de Vries et al. 2001).

This direct approach is only useful in soils where the base saturation is such that soils are in the Al buffer range. Consequently, we only applied this approach for soils with a pH less than 4.5 and base saturation below 25%. In other soils, the PDT could be estimated by calculating the needed removal in exchangeable base cation pool in addition to the critical load value. It is clear that this will result in very high values that are never exceeded by the present loads and therefore, those soils were excluded from the analyses (compare De Vries et al., 2000a).

As with N leaching, Al leaching at a given site in dependence of the acid deposition was calculated according to:

$$Al_{le} = E + F \cdot (S + N)_{dep} \text{ or } Al_{le,crit} = E + F \cdot PDT(S + N)$$

$$(4.36)$$

Again, E is a site-specific value, that is calculated by applying Eq. (4.35) and multiplying the result by the ratio of the measured and predicted Al leaching, whereas F is regression coefficient  $\alpha_n$  multiplied by the ratio of the measured and predicted Al leaching. Use of Eq. (4.36) directly leads to an estimate of PDT(S+N) according to:

$$PDT(S+N) = (Al_{le,crit} - E)/F$$
(4.37)

By using this direct empirical approach, the acid buffering by BC, S and N retention or release at the present deposition threshold is indirectly accounted for. In Eq (4.37), the value of  $Al_{le,crit}$  was derived by multiplying the present base cation leaching with a critical Al/Bc ratio. The base cation leaching at the present deposition threshold was calculated by assuming that it stays equal to the present base cation leaching.

An alternative approach that was also investigated was to calculate the base cation leaching as a function of the acid deposition according to: 100

$$BC_{le} = G + H \cdot (S + N)_{dep}$$

$$(4.38)$$

According to the previous approaches, G is a site-specific value, that is calculated by applying Eq. (4.35), with  $Bc_{le}$  as the response variable instead of  $Al_{le}$ , and multiplying the result by the ratio of the measured and predicted BC leaching and H is regression coefficient  $\alpha_n$  multiplied by the ratio of the measured and predicted Bc leaching. The differences in the response of Al and Bc to the acid deposition would then lead to a present deposition threshold where the Al/Bc ratio equals a critical value, which can be calculated according to:

$$PDT(S+N) = (Al/Bc_{crit} \cdot G - E)/(1 - Al/Bc_{crit} \cdot H - F)$$
(4.39)

Especially for acidity, the present deposition threshold can differ strongly from the long-term critical load. This can for example be due to strong present buffering by cation exchange in well buffered soils and inversely to a strong release of sulphate, associated with acid production, in soils where the present input of sulphur is much lower than in the past. In the first case a PDT can become very high, whereas in the latter case, a PDT can become negative (even at zero input, the Al concentration would not directly recover to a value at or below the critical limit).

### 4.2.4.3 Assessment of input data

Below we first describe the assessment of input data for the calculation of long-term critical acid loads and then the approach used to assess present deposition thresholds. For critical loads, we focus on the base cation inputs and outputs and the critical acidity leaching, For the N retention terms, we refer to Section 4.2.3.3.

#### Deposition of acidity and base cations

A calculation of the total deposition of sulphur for the period 1995-1998 was derived by adding measured throughfall and measured or estimated stemflow values below the forest canopy, assuming that the effects of foliar sulphur uptake by the forest canopy is negligible. For nitrogen, foliar uptake was calculated as described before (Section 4.2.3.3). Base cation (Ca, Mg, K and Na) deposition data for each stand for the period 1995-1998 was derived on the basis of through fall and bulk deposition data, accounting for canopy exchange, as described in De Vries et al. (2001).

#### Base cation weathering

There are various possibilities to assess weathering rates including (UNECE, 1996):

- Estimation of the depletion of base cations in the soil profile by chemical analyses of different soil horizons including the parent material. This method, in which an extremely resistant mineral, such as zirconium, is often used as an internal standard, gives the average weathering rate over the period of soil formation (De Vries and Breeuwsma, 1986, Starr et al., 1998).
- Correlation between the weathering rate and the total Ca and Mg content in the parent material multiplied by the present day effective temperature sum (Olsson and Melkerud, 1991). As with the previous approach, this gives the average weathering rate over the period of soil formation.
- The weathering rate model PROFILE, which calculates actual field weathering rates based on the soil mineralogy (Sverdrup, 1990; Sverdrup and Warfvinge, 1993).

- Assignment of an actual field weathering based on the parent material and texture class of a given soil (dominant) soil unit (De Vries et. al, 1994a).

Apart from the last method, total cation concentrations in either the parent material (C horizon) or even in the complete soil profile, are needed to estimate the weathering rate. At present, PROFILE is most frequently used in critical load calculations when detailed mineralogical data are available or when such data can be derived from a total cation analyses. Becker et al (2000), for example, used PROFILE to calculate the weathering of Intensive Monitoring plots in Germany for the assessment of critical loads for those plots. Starr et al. (1998) compared the weathering rates in an integrated monitoring catchment in Finland, dominated by glacifluvial sand, using the Zr depletion method, the total Ca and Mg content correlation method and the PROFILE model. Results for the sum of Ca and Mg weathering appeared to be in the same order of magnitude and varied between 130 and 280 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>.

Data on the total cation contents are, however, either not available for the plots or not submitted to FIMCI. Consequently, base cation weathering rates for the root zone were derived by a relationship with parent material class and texture class (either available or derived from soil type information), as described in Table 4.8. The estimates thus derived were updated on the basis of either the measured annual average temperature in the considered period 1995-1998 or interpolated annual average values for the 10-year period 1985-1995, using a procedure described in De Vries et al. (1994a). The weathering rates for sandy textures are in the range given by Starr et al. (1998). More information on the reliability of this approach is given in Section 4.4.1.

Parent material class	Weatherin	ng rate (mol <sub>c</sub> .ha <sup>-1</sup>	.yr <sup>-1</sup> )			
	1	1/2	1/3	2	2/3	3
Acidic <sup>1)</sup>	125	375	-	625	875	-
Intermediate <sup>2)</sup>	375	625	875	875	1125	1375
Alkaline <sup>3)</sup>	375	625	-	1125	1375	-

 Table 4.8
 Weathering rates used for the various combinations of parent material class and texture class (indicated by 1, 1/2, etc.) that occur below forests for a rootzone of 50 cm.

<sup>1)</sup> Acidic: sand (stone), gravel, granite, quartzite, gneiss (schist, shale, greywacke, glacial till). Schist, shale, greywacke and glacial till are put in brackets since soil types containing these parent materials can be converted to the acidic or intermediate parent material class, depending on the other parent materials available.

<sup>2)</sup> Intermediate: gronodiorite, loess, fluvial and marine sediment (schist, shale, greywacke, glacial till)

<sup>3)</sup> Alkaline: gabbro, basalt, dolomite, volcanic deposits.

# Base cation uptake

As with nitrogen, net base cation uptake was calculated at each site by multiplying the annual average growth rate of stems with the density and the base cation content in stems. For the derivation of those data we refer to Section 4.2.3.3, where the derivation of the net uptake of nitrogen is presented. Data on the base cation contents in stemwood are also presented in Table 4.5 in this section.

# Critical acidity leaching

The critical acidity leaching rate was calculated by multiplying the precipitation excess by a critical acidity, related to the stand still principle (no change in base saturation or readily available aluminium) or the effect based principle (a critical Al concentration or Al/(Ca+Mg+K ratio), as described above in Section 4.2.4.2. The precipitation excess was calculated with a water balance model described in De Vries et al. (2001).

### Present deposition thresholds

For the assessment of present threshold depositions, use was made of the results of input output budgets for 121 Intensive monitoring plots to get BC leaching and Al leaching. In the ultimate equation used, the climatic region, stand age, altitude and the base saturation in the mineral subsoil appeared to be significant factors in the assessment of the Al leaching rate. The coefficient relating Al leaching to N and S deposition, while accounting for the other factors described before, equalled 0.3804. The value of PDT(S+N) was then calculated with Eq (4.37) according to the procedure described above.

# 4.3 Results

A comparison of present element inputs from the atmosphere and critical loads give insight into the sites that are potentially at risk. In this section we give such information , distinguishing between critical loads based a stand-still approach and an effect-based approach. It is important to realise that the term critical load is generally limited to an effect-based approach only. An exceedance of a stand-still load only implies accumulation of nitrogen or net release of base cations or aluminium form the soil system. Specifically in the case of nitrogen, an exceedance may initially even be beneficial in N limited systems. It only indicates that the system is in transition.

# 4.3.1 Critical nitrogen loads and their exceedances

# Comparison of average present loads and critical loads for nitrogen related to N accumulation

Information on present loads, critical loads and the percentage of plots where critical loads for nitrogen are exceeded, based on the stand-still approach (no N accumulation) while distinguishing between the most important tree species in Europe, is given in Table 4.9. Values are given in  $mol_c.ha^{-1}.yr^{-1}$ . Values in kg.ha<sup>-1</sup>.yr<sup>-1</sup> can be derived by multiplying those results with a factor 14/1000.

Tree Number of			CL stand-still (mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )		CL excess (%)	
species	sites <sup>1</sup>	$(\text{mol}_{c}.\text{ha}^{-1}.\text{yr}^{-1})$	Present N leaching <sup>2</sup>	Natural N leaching <sup>3</sup>	Present N leaching <sup>2</sup>	Natural N leaching <sup>3</sup>
Pine	23 (57)	1074	704	419	91	96
Spruce	49 (96)	1359	1018	618	88	86
Oak	15 (8)	1476	1319	623	73	93
Beech	17 (42)	1540	1008	659	88	98
Other	7 (1)	1198	804	670	57	91
All	111 (234)	1329	978	580	85	92

 Table 4.9
 Average present deposition load (PDL), critical load related to accumulation of N in the present or pristine situation (CL) and percentage of plots exceeding a critical load (CL excess) for nitrogen.

<sup>1</sup> The first number refers to the plots with data on present N leaching rates and the value in brackets to the plots with calculated data on natural low N leaching rates.

<sup>2</sup> Based on present N leaching rates that were available at 111 of the 234 plots considered

<sup>3</sup> Based on a natural low N leaching rate, calculated for all 234 plots.

The average present nitrogen load on the investigated 234 plots was nearly 20 kg.ha<sup>-1</sup>.yr<sup>-1</sup>. Lowest loads were found for pine, followed by spruce, reflecting their location in mostly low deposition areas, such as Scandinavia.

Stand-still loads, which aim at no further net accumulation of nitrogen, were calculated by using the present N leaching rate. The average value thus derived was nearly 14 kg.ha<sup>-1</sup>.yr<sup>-1</sup>, and varied

mostly between 7 and 25 kg.ha<sup>-1</sup>.yr<sup>-1</sup>. At 35% of the plots, these loads were exceeded, implying N accumulation in the soil (Table 4.9). Apart from using the present N leaching rate, stand-still loads were also calculated by using a natural N leaching rate, requiring that the N concentration in the soil solution in the subsoil is as low as 0.02 mol<sub>c</sub>.m<sup>-3</sup> (approximately 0.3 mgN.l<sup>-1</sup>). Using this limits leads to a very low N leaching rate from the system and consequently to low critical nitrogen N loads, which ranged mostly between 2 and 14 kg.ha<sup>-1</sup>.yr<sup>-1</sup> with an average near 8 kg.ha<sup>-1</sup>.yr<sup>-1</sup>. Values were clearly lower for pine, with a lower N uptake, than for the other tree species. This critical N load was exceeded at 92% of the plots (Table 4.9). The limit of 0.02 mol<sub>c</sub>.m<sup>-3</sup> has also been suggested in literature in relation to possible impacts on the species composition of the ground vegetation. It is however not correct to suggest that at 92% of the plots the composition of the ground vegetation will change because of too high N inputs, since the calculated critical loads are lower than empirical data, which vary mostly between 7 and 20 kg.ha<sup>-1</sup>.yr<sup>-1</sup> with an average near 14 kg.ha<sup>-1</sup>.yr<sup>-1</sup> (see Section 4.2.2.1).

# Comparison of average present loads and critical loads for nitrogen related to impacts on ground vegetation and trees

Information on effect-based critical loads and the percentage of plots in which present loads, exceed those critical loads, distinguishing between the most important tree species in Europe, is given in Table 4.8. A distinction is made between impacts on ground vegetation and coniferous trees. The critical loads related to impacts on ground vegetation, based on a critical N concentration limit of 0.2 mol<sub>c</sub>.m<sup>-3</sup> (approximately 3 mgN.l<sup>-1</sup>), leads to an average critical load of 17 kg.ha<sup>-1</sup>.yr<sup>-1</sup> and a median value of 13 kg.ha<sup>-1</sup>.yr<sup>-1</sup>. These loads are exceeded at 58% of the plots. This comparison shows that changes in plant biodiversity are likely in large parts of the European forests. Effect–based critical loads which aim at concentrations of nitrogen in the foliage below a critical limit were only calculated for conifers, using a critical limit of 1.8 g.kg<sup>-1</sup>. Above this limit the risk for drought stress, frost, pest and diseases increases. Such limits are not known for oak and beech and furthermore, the relation between N deposition and foliar N concentration is not so clear for these deciduous trees. The critical load thus calculated was near 14 kg.ha<sup>-1</sup>.yr<sup>-1</sup> for pine and near 20 kg.ha<sup>-1</sup>.yr<sup>-1</sup> for spruce. These critical loads were exceeded at 45% of the plots with coniferous trees (Table 4.10).

Tree	Number of	PDL	CL effect- based (mol	<sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )	CL excess (%)	
species	sites	$(mol_c.ha^{-1}.yr^{-1})$	Ground vegetation <sup>1)</sup>	Trees <sup>2)</sup>	Ground vegetation <sup>1)</sup>	Trees <sup>2)</sup>
Pine	57	1074	901	1006	59	53
Spruce	96	1359	1323	1463	47	44
Oak	28	1476	1182	-	75	-
Beech	42	1540	1257	-	81	-
Other	11	1198	1874	1534	17	20
All	234	1329	1219	1308	58	45

 Table 4.10
 Average present deposition load (PDL), critical load related to impacts on ground vegetation and trees (CL) and percentage of plots exceeding a critical load (CL excess) for nitrogen.

<sup>1)</sup> Results based on an N leaching rate that is related to impacts on the species composition of the ground vegetation.

<sup>2)</sup> Results based on an N leaching rate that is related to critical N concentrations in the foliage of conifers. This critical load could thus not be calculated for oak and beech.

The results based on limits for tree impacts are in reasonable agreement with those presented in De Vries et al. (2000a), who used a relationship found by Tietema and Beier (1995) between N concentrations in foliage and N leaching rates for a number of intensively monitored plots (NITREX sites). Applying the critical N concentration in this relationship lead to a critical N leaching rate of 20 kg.ha<sup>-1</sup>.yr<sup>-1</sup> and critical loads that are slightly above this value. The results are also comparable to empirical critical loads based on a relationships between atmospheric N 104

deposition and N content in foliage. Figure 4.6 gives an example of such a relationship for Scots pine and Norway Spruce at 68 intensive monitoring plots in Europe. Nearly 70% of the variation in foliar N concentration of Scots pine could be explained by atmospheric deposition, using a non-linear relationship. Using this relationship, a critical N concentration in the needles of 18 g.kg<sup>-1</sup> was reached near a deposition level of 20 kg.ha<sup>-1</sup>·yr<sup>-1</sup> for Scots pine (Fig. 4.6), being comparable to the average value derived with model calculations. Both critical loads are in the range given in Table 4.2 for tree health. For Norway spruce, the critical limit is hardly ever exceeded.

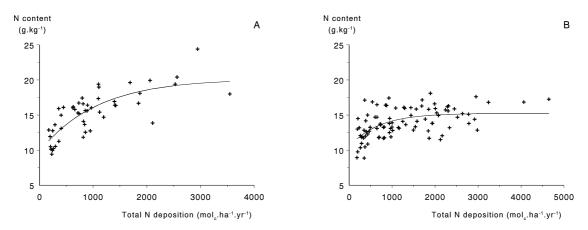


Figure 4.6 Relationship between nitrogen contents in first year needles of Scots pine (A) and Norway spruce (B) and total nitrogen deposition at 68 plots in Europe, showing the possibility to set an empirical critical nitrogen load.

# Geographic variation in critical nitrogen loads and their exceedances

The geographic variation in critical nitrogen loads related to nitrogen accumulation, assuming a natural N leaching rate (acceptable N concentration in soil solution of 0.02 mol<sub>c</sub>.m<sup>-3</sup>) and their exceedances by present loads are presented in Fig. 4.7. Results show that high critical nitrogen loads (>1000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>) only occur in Southern Europe, mainly due to high N uptake, specifically by broadleaves. Critical nitrogen loads were calculated to be much smaller in northern Europe, where the net uptake of N by trees is low. The geographic variation in the exceedance of critical nitrogen loads is large. Highest exceedances in view of possible N accumulation (acceptable N concentration in soil solution of 0.2 mol<sub>c</sub>.m<sup>-3</sup>) do occur in Western and Central Europe where present loads of nitrogen are generally high and critical loads are relatively low. In the Nordic countries, the excess is generally less than 400 molc.ha<sup>-1</sup>.yr<sup>-1</sup>. Figure 4.8 and 4.9 present the geographic variation in critical nitrogen loads and their exceedances related to impacts on ground vegetation and tree nutrition of conifers (acceptable N concentration in needles is 1.8%), respectively. The results show that effect-based critical loads are mainly exceeded in Central and Western Europe and hardly in Northern Europe. It should also be kept in mind that this exceedance does not necessarily mean that critical N concentrations or critical N contents in foliage are presently exceeded at those plots. It only means that in the long term, in a steady-state situation, such an exceedance is likely to occur at continuing present nitrogen loads. An evaluation of the present situation requires the calculation of present deposition thresholds for nitrogen, which are discussed in Section 4.3.3.

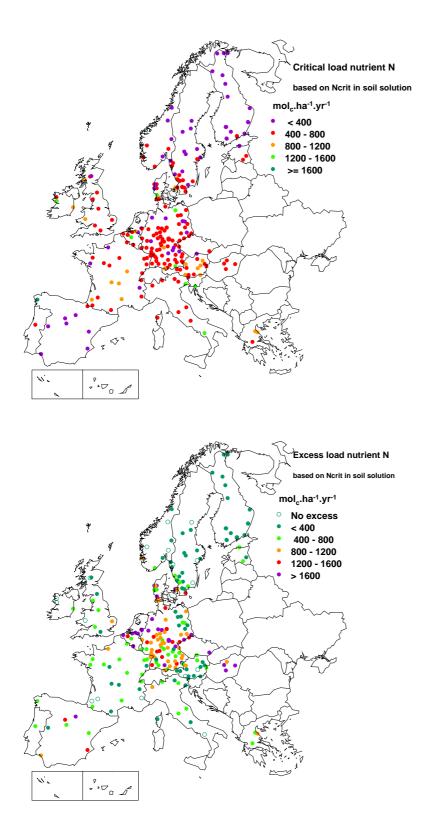


Figure 4.7 Geographical variation of critical loads (top) and critical load exceedances (bottom) for nitrogen  $(mol_c.ha^{-1}.yr^{-1})$  based on the stand-still approach (no nitrogen accumulation starting from a pristine situation assuming a natural N concentration in soil solution of 0.02 mol<sub>c</sub>·m<sup>-3</sup>)

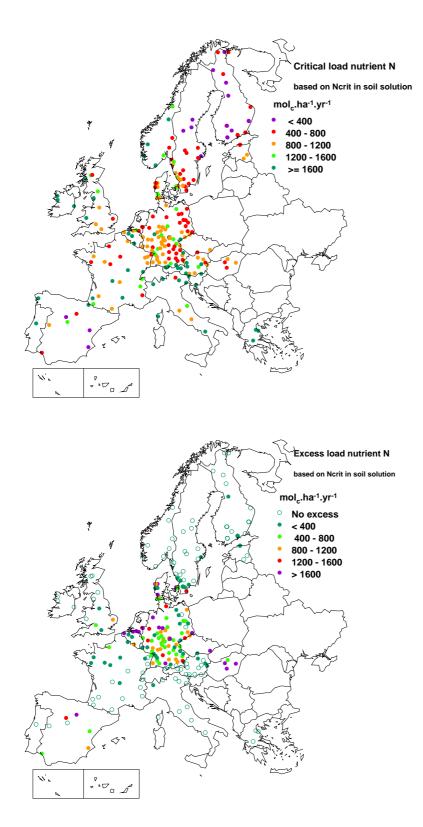


Figure 4.8 Geographical variation of critical loads (top) and critical load exceedances (bottom) for nitrogen  $(mol_c.ha^{-1}.yr^{-1})$  using an effect-based approach related to impacts on ground vegetation (acceptable N concentration in soil solution is  $0.2 \text{ mol}_c.m^{-3}$ )

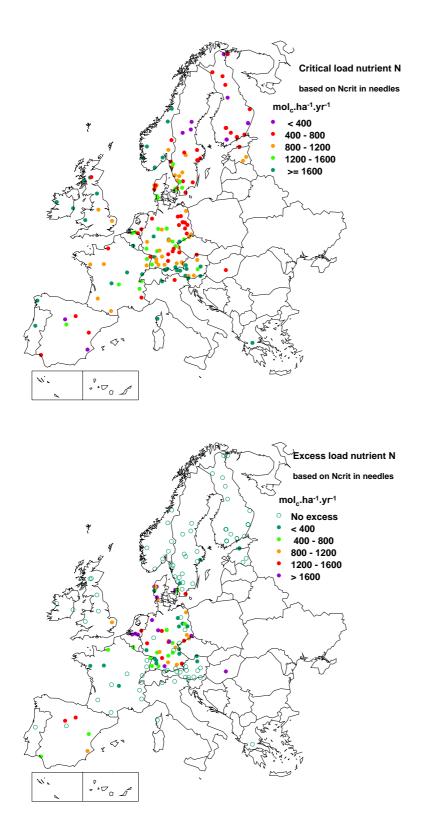


Figure 4.9 Geographical variation of critical loads (top) and critical load exceedances (bottom) for nitrogen  $(mol_c.ha^{-1}.yr^{-1})$  using an effect-based approach related to impacts on tree nutrition of conifers (acceptable N concentration in needles is 1.8%).

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### 4.3.2 Critical acid loads and their exceedances

#### Comparison of average present loads and critical loads related to impacts on soil

Information on present loads, stand-still loads and the percentage of plots exceeding critical loads for acidity for the most important tree species in Europe is given in Table 4.11. Critical loads were calculated by requiring no further loss of exchangeable base cations in base rich forests soil (loess, clay and peat soils) and no further loss of readily available aluminium in base poor sandy forest soils (Stand-still principle).

 Table 4.11
 Average present deposition load (PDL), critical load (CL) and percentage of plots exceeding a critical load (CL excess) for acidity related to impacts on soil

Tree	Number of	PDL	CL stand-stil	l (mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )	CL excess (%)	
species	sites	(mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )	Standard <sup>1)</sup>	Alternative <sup>1)</sup>	Standard <sup>1)</sup>	Alternative <sup>1)</sup>
Pine	57	1749	1724	1755	45	44
Spruce	96	2146	1487	1499	65	62
Oak	28	2272	1642	1650	74	74
Beech	42	2346	1643	1638	80	83
Other	11	2032	2299	2324	60	60
All	226	2094	1627	1640	64	62

<sup>T</sup>) Standard implies the use of a standard gibbsite coefficient, whereas alternative implies the use of optimised coefficients per soil type

The average present acid load on all investigated 226 plots was nearly 2100 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. As with nitrogen, lowest loads were found for pine, followed by spruce. The stand-still acid loads were on average approximately 1600 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>, and were exceeded at approximately 60-65% of the plots independent of the method used. The use of the standard gibbsite coefficient, used in most model calculations up to now does not seem to deviate much from the use of different Al release constants per soil type. This shows that the method in this respect is rather robust.

#### Comparison of average present loads and critical loads for acidity related to impacts on trees

Information on effect-based critical loads and the percentage of plots exceeding those critical loads for acidity for the most important tree species in Europe is given in Table 4.12.

 Table 4.12
 Average present deposition load (PDL), critical load (CL) and percentage of plots exceeding a critical load (CL excess) for acidity related to impacts on trees

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Tree	Number of PDL		CL effect-ba	sed (mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )	CL excess (%)	)
species	sites	(mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )	Standard <sup>1)</sup>	Alternative <sup>1)</sup>	Standard <sup>1)</sup>	Alternative <sup>1)</sup>
Pine	57	1749	2906	2995	40	30
Spruce	96	2146	2726	2998	34	35
Oak	28	2272	4721	4508	25	25
Beech	42	2346	4624	4577	31	26
Other	11	2032	5282	5180	18	18
All	226	2094	3469	3564	33	30

<sup>1)</sup> Standard implies the use of a standard gibbsite coefficient and critical Al/Bc ratios only, whereas alternative implies the use of optimised coefficients per soil type and the additional use of a critical Al concentration in situations where the Bc concentration is very low, thus avoiding too low critical limits for the Al concentration.

The effect–based critical loads were calculated by aiming that ratios of toxic aluminium to base cations in the soil solution stay below a critical limit of 0.8 for pine and spruce and 1.6 for oak and beech. These critical acid loads were approximately twice as high as the stand-still loads. Values ranged mostly between 500 and 8000, with an average near 3500 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. As with the stand-still loads, the results of effect-based critical acid loads were nearly independent of the

Al release constants used (Table 4.9) Results are clearly lower for pine and spruce, which are more sensitive to aluminium, than for oak and beech. Considering all plots, critical loads were exceeded at 30-33% of the plots when impacts on tree roots are considered This is in line with measurements of aluminium to base cations ratios in the soil solution and shows that impacts on forests are likely in a substantial part of the European forests.

### Geographic variation in critical acid loads and their exceedances

The results related to the stand-still approach (no further depletion of base cations or readily available aluminium) show a comparable pattern although the critical loads are lower and the critical load exceedances are higher (Fig. 4.10). The geographic variation in critical loads and critical load exceedances for acidity related to impacts on tree roots (acceptable molar aluminium to base cation ratio of 0.8 for conifers and 1.6 for broadleaves) are shown in Fig.4.11. The results show that high critical acid loads (>3000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>) mainly occur in Southern Europe due to high weathering rates and the more preferent occurrence of broadleaves, which are less sensitive to acidification. In general, the critical acid load increased from the northern boreal regions to Southern Europe, due to an increase in base cation input from the atmosphere and by soil weathering and in nitrogen uptake. As with nitrogen, the geographic variation in the exceedance of critical loads is large with exceedances in Western and Central Europe, where present loads of acidity are generally high and critical loads are relatively low. In the remaining plots, the exceedances are generally negligible, except for the Southern part of Scandinavia where low exceedances do occur up to 600 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. It should be kept in mind, however, that this exceedance does not necessarily imply that critical Al/Bc ratios or Al concentrations are presently exceeded at those plots. It only means that in the long term, in a steady-state situation, such an exceedance is likely to occur at continuing present acid loads. As with the results for nitrogen, an evaluation of the present situation requires the calculation of present deposition thresholds for acidity, which are discussed below in Section 4.3.3.

## 4.3.3 Present deposition thresholds for nitrogen and acidity and their exceedances

Calculated present deposition thresholds (PDT) for nitrogen and acidity often strongly deviated from the long-term critical loads (CL). In plots where the present concentrations of e.g. N and Al in soil solution exceeded the critical limits, PDT was lower than CL and often became negative (it is impossible to attain a critical limit within one year starting with the present high Al or N concentration). At plots with the present Al or N concentrations below critical limits, the PDT was generally much larger than the CL. Negative values were quite often encountered for the PDT of acidity, implying that the critical limit can only be attained by liming if one wants to attain the critical limit in a one-year period. Inversely, extreme high values for PDT acidity were found for well buffered soil, where the pool of exchangeable base cations prevents soil acidification. Even though this aspect was already partly accounted for by only including acidic plots, extremely high values were sometimes calculated. In policy making, the term target load (TL) is also used in which a longer time period is defined, e.g. 100 years. Also for these target loads, the general principle holds that TL is lower than CL if the present concentrations exceed critical limits, whereas in the opposite situation, TL is larger than CL, but the differences are much less extreme than those between PDT and CL.

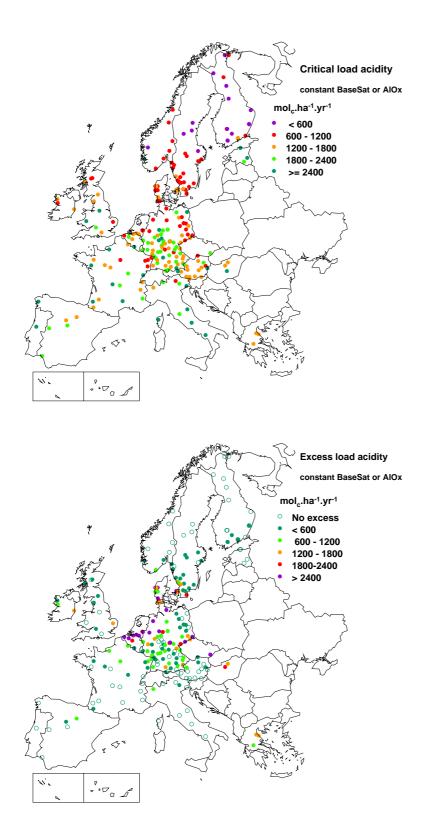
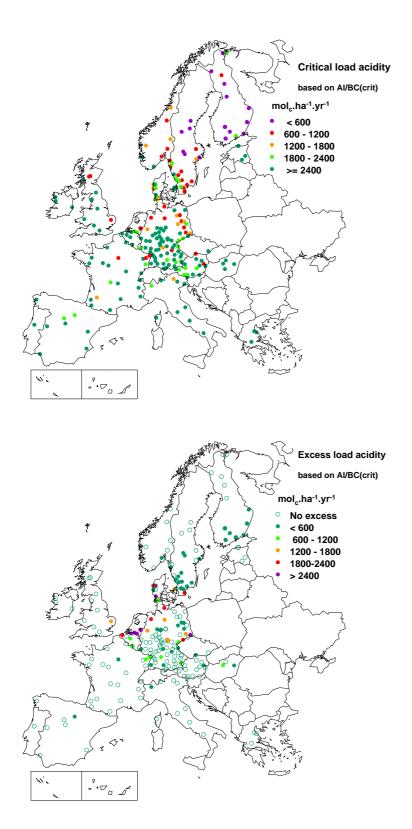


Figure 4.10 Geographical variation of critical loads (top) and critical load exceedances (bottom) for acidity  $(mol_c ha^{-1}.yr^{-1})$  using the stand-still approach (no further depletion of base cations or readily available aluminium).



*Figure 4.11 Geographical variation of critical loads (top) and critical load exceedances (bottom) for acidity (mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>) using an effect–based approach related to impacts on tree roots (acceptable molar aluminium to base cation ratio of 0.8 for conifers and 1.6 for broadleaves).* 

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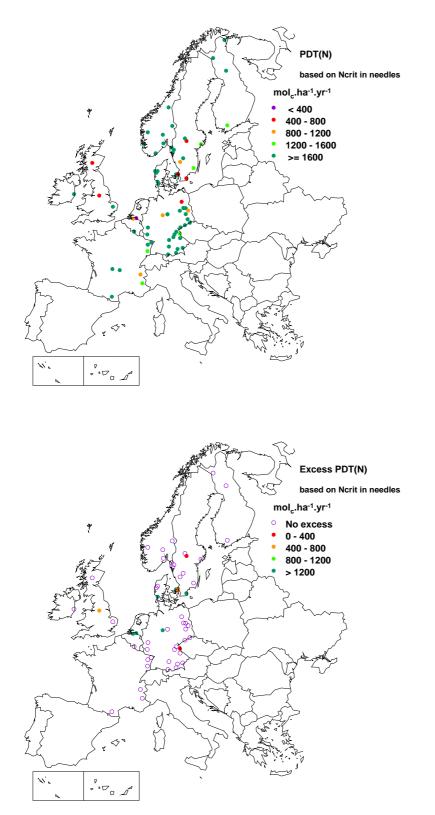


Figure 412 Geographical variation of present deposition thresholds (top) and their exceedances by present loads (bottom) for nitrogen  $(mol_c.ha^{-1}.yr^{-1})$  related to impacts on tree nutrition of conifers (acceptable N concentration in needles is 1.8%).

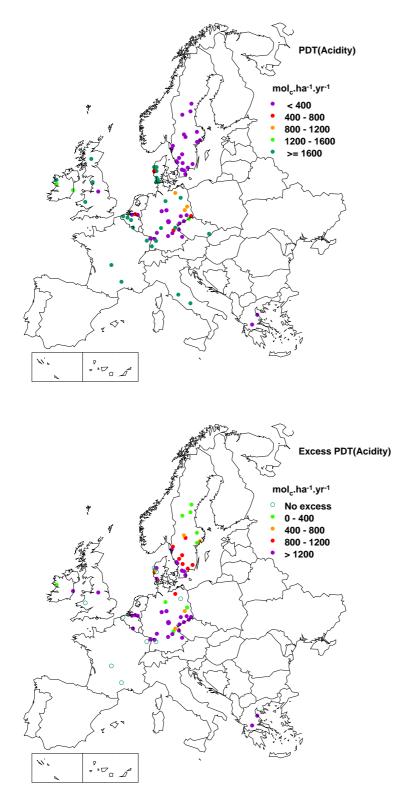


Figure 4.13 Geographical variation of present deposition thresholds (top) and their exceedances by present loads (bottom) for nitrogen ( $mol_c$ · $ha^{-1}$ · $yr^{-1}$ ) related impacts on tree roots (acceptable molar Al/BC ratio is 0.8 for conifers and 1.6 for broadleaves).

Considering the aspects above, the presentation of mean values for PDT's was considered irrelevant. Instead we only present the geographic patterns of PDT's and their excesses, focusing on the tree compartment by using the N contents in foliage and the Al/Bc ratio in the soil solution as critical limits. These patterns do give an indication where forests are presently at risk in view of high atmospheric loads. Fig. 4.12 presents the PDT's related to tree impacts, in terms of elevated N concentrations in the foliage of conifers (results are thus limited to these tree species), and their exceedances by present loads. A comparison of the results with those obtained for critical loads shows that the present deposition thresholds are generally much larger than the steady-state critical loads and consequently the exceedance is much lower (Compare Fig. 4.12 and 4.9). The calculated exceedance of the PDT for the 72 considered plots was only 15%, compared to 45% of the 164 considered plots in the CL calculation (Table 4.10).

Fig. 4.13 presents the PDT's related to tree root impacts, in terms of elevated dissolved Al/Bc ratios, and their exceedances by present loads. Unlike nitrogen, the PDT's for acidity related to those criteria were generally lower than the CL values. Consequently, the area exceeding the critical loads was higher. One has to consider, however, that the results were limited to 84 plots with low pH or elevated Al leaching values only. For all other plots the present deposition is certainly lower than the present deposition threshold. In Finland for examples, no calculations were made considering this prerequisite (Compare Fig 4.13 and 4.11). Even when one considers this aspect, it nevertheless implies that the present situation may be even worse than the steady state situation which is most likely due to net release of sulphur in many plots with previous high sulphur inputs (see the previous Technical Report; de Vries et al., 2001). The relatively large exceedance of critical Al/Bc ratios, up to 46%, was also presented in previous reports (De Vries et al., 2000b), being higher than the calculated percentage of plots exceeding long-term critical loads (33%). The results confirm the present non steady state situation for nitrogen in terms of N accumulation and for acidity in terms of acidity (sulphur) release.

## 4.4 Discussion and conclusions

## 4.4.1 Uncertainties in critical loads

### Limitations of the steady-state model approach

In this study, steady-state soil models were used to calculate critical loads because they are simple and relatively easily applicable. This is also the case with empirical data but an empirical approach can only be applied to derive critical N loads, due to the large influence of N on the species diversity of terrestrial ecosystems. A major drawback of steady-state models, and even of simple dynamic soil models, is the neglection of biotic interactions. For example, vegetation changes in heathlands are mainly triggered by a change in N cycling (N mineralisation; Berendse et al., 1987). Neglecting biotic interactions also limits the derivation of critical N loads related to forest damage. The derivation of critical loads based on a critical foliar N concentration (see Table 3) is only possible with a steady-state soil model by using purely empirical relationships between N concentrations in the plant and in the soil solution. At present there are several integrated forest-soil models that are potentially useful for a more scientifically based derivation of critical N loads. Examples are the models NAP (Van Oene, 1992) and ForSVA (Oja et al., 1995) and SMART-MOVE (Kros et al., 1995; Latour and Reiling, 1993). The models NAP (Van Oene, 1992) and FORSVA (Oja et al., 1995) have been used to derive critical loads for acidity (the sum of N and S deposition) for a Norway spruce stand in Solling (Germany). The criteria that were used to derive a critical load were: (i) optimal growth during a relation period (100 years) while avoiding Mg deficiency (NAP model) and (ii) a long-term sustainable biomass production avoiding toxic Al effects (ForSVA model). The critical loads for the sum of N and S thus derived were close to those derived by a steady-state soil model. More information on this comparison is given in De Vries et al. (1995).

The SMART-MOVE model predicts the occurrence probability of plant species in response to scenarios for acidification, eutrophication and desiccation and consists of a soil module (SMART; De Vries et al., 1989) and a vegetation module (MOVE; Latour and Reiling, 1993). The SMART model predicts changes in abiotic soil factors indicating acidification (pH), eutrophication (N availability) and desiccation (moisture content) in response to scenarios for acid deposition and groundwater abstraction. The model MOVE predicts the occurrence of species. Since the speciesresponse functions are based on Ellenberg indicator values, a calibration of these indication values to quantitative values of the abiotic soil factors is necessary to link the soil module to the vegetation module (see Section 4.2.2.1). The advantage of SMART-MOVE is that the two approaches to assess critical loads, i.e. (i) the exceedance of critical values for ion concentrations and ion ratios in soil water, and (ii) the analysis of changes in species composition, are combined in a consistent framework. Recently, Van Hinsberg and Kros (1999) derived critical loads for nitrogen and acidity related to species diversity for the most common terrestrial ecosystems in the Netherlands with the SMART-MOVE model. Results are described in Albers et al. (2001). In general critical nitrogen loads and critical acid loads varied mostly between 500-2000 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. Furthermore, the results obtained with SMART-MOVE were within the range of empirical critical N loads.

A disadvantage of relatively complex integrated soil vegetation models is that input data for their application is generally incomplete and values can only be roughly estimated. Even if the model structure is correct (or at least adequately representing current knowledge), the uncertainty in the output of complex models may still be large because of the uncertainty of input data. Simpler empirical models have the advantage of a smaller need for input data but the theoretical basis, that is needed to establish confidence in the predictions, is small, which limits the application of such models for different situations. There is thus a trade off between model complexity (reliability) and applicability. The most relevant use of integrated dynamic soil-vegetation models, including interactions of drought, acidification and eutrophication on forests, and preferably also the effects of pests and diseases, is to check the results of conventional methods as done in the modelling study in Solling described above. An overview of the advantages and disadvantages of the various methods is given in Table 4.13. A dynamic modelling approach limited to the soil compartment is foreseen in next years report for nitrogen and acidity

Method	Advantage	Disadvantage
Empirical data	Based on empirical relationships between	Not applicable for acidity and metals (no clear
	effects and atmospheric (N) deposition	empirical relationship)
	Easily applicable	Other effects than acidification and
		eutrophication may be involved
Steady-state soil	Simple	No biotic interactions involved
Models	Easily applicable	Critical limits are uncertain
Integrated soil-	Comprehensive description of the ecosystem	Model complexity
vegetation models	Important tool to assess target loads and	Not easily applicable for mapping
-	evaluate scenarios	·

 Table 4.13. Evaluation of various methods for the assessment of critical loads

### Uncertainties in critical limits, model structure and input data

Uncertainties in critical loads derived by steady-state soil models are determined by the uncertainty in critical limits, model structure and input data. A systematic overview of these effects is given below.

### Uncertainties in critical limits

Nitrogen: The choice of the critical N leaching rate strongly affects the critical deposition levels of N. The natural N concentration in leachate of 0.02 mol<sub>c</sub>.m<sup>-3</sup> or 0.28 g.m<sup>-3</sup>, leading to N leaching rates of approximately 0.5-2 kg.ha<sup>-1</sup>.yr<sup>-1</sup> for precipitation excesses varying between approximately 200 and 800 mm.yr<sup>-1</sup> is only a proxy for the stand-still load. The value is based on annual average natural nitrate concentrations in stream water. The occurrence of NH<sub>4</sub> and organic N in stream water is neglected in this context, implying that it is probably an underestimate. Even when using natural rates, one has to be aware that there is considerable temporal variation and hydrological events may strongly affect these average estimates. Increased nitrate concentrations can occur in runoff outside the growing season, when hydrological events such as storms and snowmelt are common and biological activity is low (e.g. Mulder et al., 1997). Studies of the Gårdsjön catchment during the NITREX experiment indicated that most N loss from the catchment occurred during a few episodes (Moldan & Wright, 1998). During periods of high flow, runoff derived directly from precipitation or with contact only with the topsoil can have relatively high nitrate concentrations (Hagedorn et al., 2001). High nitrate concentrations in stream water have also been observed after summer drought (Lydersen, 1995), but these are unimportant in terms of fluxes because of low runoff. Finally, disturbances to the forest ecosystem such as clear-cuts can also temporarily raise nitrate and ammonium concentrations (Mulder et al., 1997; Hu, 2000), but this aspect has not been considered in the critical load calculations.

The use of a relatively high critical N concentrations when one wants to calculate critical N loads related to vegetation changes (e.g.  $0.2 \text{ mol}_{e}\text{.m}^{-3}$  or  $2.8 \text{ g.m}^{-3}$  as used in this study) seems more appropriate than the use of natural N concentrations, but the value is highly uncertain. One can only say that the results are plausible in view of empirical data. The critical limit for a foliar N concentration of 18 g.kg<sup>-1</sup> related to effects on trees is also prone to uncertainty. The literature cited indicates an N concentration range of 16 to 20 g kg<sup>-1</sup> in needles as optimal, whereas a review made by Morrison (1974) gives optimal concentrations between 15 to 18 g kg<sup>-1</sup>. Even though negative effects are not likely to occur below 18 g kg<sup>-1</sup>, there will be a clear range depending on other factors as well, such as the availability of base cations. In general, the uncertainties in critical limits may cause an uncertainty of at least 25% in the resulting critical loads.

*Acidity:* An uncritical use of the Ca/Al ratio has been criticised by several authors (Högberg and Jensén, 1994, Løkke et al., 1996; Binkley and Högberg, 1997). Uncertainties in critical values for the Al/(Ca+Mg+K) ratio, related to direct toxic effects of Al, are mainly due to a lack of knowledge about the effects of Al in the field situation. Although Cronan and Grigal (1995) found evidence in support of use of the Ca/Al ratio, much of the data reviewed by them was from hydroponic or soil experiments with seedlings under laboratory conditions. It is uncertain to what extent results obtained from laboratory experiments can be applied to natural forest ecosystems with adult trees, mycorrhizae etc. A forest manipulation experiment at Nordmoen (north of Oslo) has shown no support for the use of the Ca/Al ratio as an indicator for risk of forest decline (De Wit, 2000, De Wit et al., 2001). Recently, Jentschke et al. (2001) found evidence supporting the use of a fine root molar Ca/Al ratio of 0.2 in Norway spruce stands in Germany, so it may be the Ca/Al ratio in the roots rather than in the surrounding soil or soil water that is important. There is then the question of how to relate concentrations in roots to those in soil or soil water.

The uncertainty is also partly due to a natural range in the sensitivity of various tree species for Al toxicity (Sverdrup and Warfvinge, 1993). It is furthermore clear that factors such as aluminium speciation have to be taken into account, as organically complexed Al is normally far less toxic than Al<sup>3+</sup> or the monomeric cationic hydroxo complexes. The calculated Al concentration is in principle, however free Al<sup>3+</sup>. Note also that critical annual average values are generally used whereas the short-term variation can be large, with peak values in the summer. Furthermore, the critical limits are applied at the bottom of the rootzone. At lower soil depths, the critical acid load generally increases strongly, since release on inorganic Al is limited (De Vries et al., 1994b). In the organic layer, where many of the roots are and from which trees take up much of their nutrients, most dissolved Al is even organically bound (e.g. Solberg et al., 2001). Finally, the Al/(Ca+Mg+K) ratio is probably irrelevant for peat soils, since Al mobilisation hardly occurs in these soils.

If one wants to avoid a decrease in base saturation (or pH), the present base saturation has to be used in the critical load calculations. The only uncertainty in this value is the spatial variability in the field situation. The uncertainty in critical loads related to a required constant pool of readily available Al compounds is mainly due to an uncertainty in the weathering rate. In general, the critical loads based on the stand-still approach are lower than the effect-based critical loads and are more reliable.

## Model assumptions

*Nitrogen:* Uncertainties related to the description of N dynamics in the steady-state model result from (i) neglecting N fixation which is important for trees such as red alder, (ii) neglecting N adsorption although NH<sub>4</sub> fixation may play a role in clay soils and adsorption of dissolved organic nitrogen (DON), being the dominant form in low deposition areas, often occurs in podzol B horizons, (iii) assuming that nitrification is complete, while it is likely to be inhibited at high C/N ratios, (iv) the simple description of net N immobilisation, and (v) neglecting the interaction between net N uptake and a change in soil conditions (De Vries, 1994a). Even though the dynamics of the N transformation processes are strongly simplified, the resulting fluxes for net N uptake, N accumulation and denitrification seem plausible in view of available data on these processes for forest soils (De Vries et al., 1994a).

Acidity: An important assumption in the SMB model is the assumed homogeneity of the rootzone both in a horizontal and vertical direction. Use of a one-layer model implies that the critical

Al/(Ca+Mg+K) ratio refers to the situation at the bottom of the rootzone, whereas most roots occur in the topsoil. Values for the Al/(Ca+Mg+K) ratio generally increase with depth due to Al mobilisation, BC uptake and transpiration. Other assumptions in the one-layer model such as (i) disregarding sulphate interactions, (ii) neglecting complexation of Al with inorganic and organic anions and (iii) a simple hydrology, are probably less significant (De Vries, 1994).

### Input data

*Nitrogen:* When the uncertainty in the input data is known, the effect on the resulting critical N load can directly be quantified. At the intensive monitoring plots, the uncertainty in net N uptake and critical N leaching is likely to be within 25%, since site specific data are available to calculate the average annual growth rate and the precipitation excess. For N uptake, this guess may be an underestimate since the yield data are average values in intervals of 5 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup>. Estimates for denitrification and long-term immobilisation, are, however, less certain and the uncertainty is likely to be larger than 50% resulting in an average uncertainty in critical N loads near 50% (see also De Vries et al.,1994a).

*Acidity:* Assuming that the model structure is correct, the effect of the uncertainty in the input data can directly be quantified. The uncertainty in the calculated net base cation input (deposition and weathering minus uptake), combined with the uncertainty in Al/(Ca+Mg+K) ratio, which affects the associated acidity leaching, has the largest effect on the calculated critical acid deposition level. One important point is that the weathering rate has been estimated from the parent material only. An impression of the reliability of the weathering rates obtained by this soil assignment method can be derived from a comparison with results obtained with the validated PROFILE model for Intensive Monitoring plots in Germany (Becker et al., 2000). These results indicate a reasonable reliability, assuming that the PROFILE approach does give an adequate estimate of the weathering rate. Considering the above mentioned uncertainties in input data, it is likely that the overall uncertainty in critical acid deposition levels varies mostly between plus or minus 50%.

## 4.4.2 Conclusions

In general, one can conclude that steady-state soil models do form a relevant tool to calculate critical loads, being a measure of possible adverse effects on terrestrial ecosystems. Indications for such effects become stronger when they are supported by empirical data (specifically the case for nitrogen) or by calculations with integrated soil-vegetation models. In general, the critical loads increase going from impacts on soil to impacts on the forest ecosystem (ground vegetation and trees). This is illustrated in a summarising table of the results obtained (Table 4.14).

 Table 4.14
 Average critical loads (CL) and percentage of plots exceeding a critical load (CL excess) for nitrogen and acidity depending on the receptor considered.

Effect considered	CL (mol <sub>c</sub> .ha <sup>-1</sup> .yr <sup>-1</sup> )	CL excess (%)			
	Nitrogen	Acidity	Nitrogen	Acidity	
Soil	580 <sup>1</sup>	1627	92	64	
Ground vegetation	1219	_2	58	_2	
Trees	1308	3469	45	33	

<sup>1</sup>Assuming a pristine situation. Critical loads related to N accumulation that were based on present N leaching at 111 plots lead to an average critical load of 978 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>.

<sup>2</sup> For acidity, no calculations were made related to impacts on ground vegetation.

Considering the limitations of the critical load approach mentioned above, the following conclusions can be drawn:

- At approximately 50% of the investigated plots, critical nitrogen loads related to impacts on ground vegetation and on the vitality of coniferous trees are exceeded by the present deposition. At these plots the risk for drought stress, frost, pests and diseases is increased and additionally the species diversity of the ground vegetation might be endangered. At approximately 90% of the plots, it is likely that N is accumulating in the ecosystem. This conclusion holds for the evaluated 234 plots, which mainly occur in Central Europe with an average nitrogen deposition of 19 kg.ha<sup>-1</sup>.yr<sup>-1</sup>.
- The average acid (nitrogen plus sulphate) deposition on the evaluated 226 plots is approximately 2100 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. At approximately two thirds of the investigated plots, the critical acid loads for soil are exceeded, implying a net loss of nutrient base cations or readily available aluminium from the soil. At approximately two thirds of the investigated plots, the critical acid loads related to the functioning of tree roots are exceeded. This conclusion holds for the evaluated 226 plots, which mainly occur in Central Europe with an average acid (nitrogen plus sulphate) deposition of approximately 2100 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. Those plots do encounter a risk for tree impacts, such as increased defoliation and decreased growth.
- Highest exceedances of critical loads do occur in central Europe, and for nitrogen also in western Europe, where present loads are high and critical loads are relatively low.

# 5 Critical loads for heavy metals and their exceedances

# **5.1 Introduction**

### Use of the critical load concept in science and policy with respect to heavy metals

Critical loads for heavy metals are of a more recent development than those for nitrogen and acidity. After a pilot study for forests in Europe (Reinds et al., 1995), a draft manual for the calculation and mapping of heavy metals (De Vries and Bakker, 1996) was discussed and amended at a UN/ECE workshop in Bad Harzburg (Germany). To date, updated manuals for terrestrial ecosystems (De Vries and Bakker, 1998) and aquatic ecosystems (De Vries et al., 1998) are available. A summary of the approaches has recently been made in a short guidance for the calculation of critical metal loads (De Vries et al., 2001). The manuals and guidance have been used by several European countries to calculate and map critical loads for lead (Pb) and cadmium(Cd) in support of European pollution reduction policy in response to a call by the ICP on mapping and modelling at the end of 2001. The approach has, however, not yet been used in policy making. The recent (1998) protocol on heavy metal emission abatement under the LRTAP Convention is based on flat rate reductions using best available abatement techniques, ignoring differences in susceptibility of receptors to metal input. It is not yet clear whether the critical load approach will be the next step in abating heavy metal emissions under the Convention. At present, there is no guarantee that it will lead to more cost-efficient emission reductions, considering the uncertainties in the critical load assessments.

### Differences between critical loads and present deposition thresholds

As with nitrogen and acidity, critical metal loads are calculated with steady-state soil models based on a simple mass balance approach. These models calculate deposition loads that avoid the violation of a critical limit for metals in soil or soil solution in a steady-state situation. Accumulation of metals in soil by adsorption, that may play a role on a time scale of several hundreds of years, is not accounted for. Adsorption processes are only included to calculate critical dissolved metal concentrations from critical soil metal concentrations. A comparison of present loads and present deposition thresholds, that lead to metal concentrations in soil solution that are equal to critical limits at present, would give more information on the ecosystems that are at risk. Plots for which present deposition thresholds could be calculated were, however, limited to those with information on bulk deposition, throughfall, metal concentrations in soil and soil solution and meteorological data to allow the calculation of metal deposition and metal leaching and thus the retention or release of metals. Such budgets, which have for example been made for a few ICP IM sites (Ukonmaanaho and Starr, 2001), based heavy metal concentrations in various aqueous and biotic media (Ukonmaanaho et al., 1998), could be calculated for very few Intensive Monitoring plots only. Furthmermore, present metal retention rates should be scaled to calculate the present deposition threshold (see the Sections 4.2.3.2 and 4.2.4.2 for nitrogen and acidity, respectively) and such a scaling can not be calculated independent of acidity status of the soils and its possible change by acidic deposition. Short-term changes in dissolved metal concentrations are much more related to the immediate effect of pH changes than to the long-term accumulation of metals. Considering these aspects, the calculation of a present deposition threshold was not considered relevant for metals. However, in evaluating the results, it is crucial than one realises the limited actual value of a long-term critical load in view of what has been

mentioned above. The results should thus be considered as preliminary and tentative (see also Section 5.4.1).

# Contents of this chapter

Up to now, calculations of critical metal loads have been made by 11 European countries, based on estimated average data on tree uptake, soil weathering and critical metal leaching. Calculations of critical metal loads and their exceedances have not yet been made for Intensive Monitoring plots and this report presents such an assessment for 245 Intensive Monitoring plots in where data on hydrology and/or soil chemistry were available. Since metal deposition data were hardly available, use was made of modelled data to get an indication of the possible exceedance. The metals that were evaluated are lead (Pb) and cadmium (Cd), being priority metals for which a call has been issued by ICP M and M to perform critical load calculations, and copper (Cu) and zinc (Zn), which are important micro nutrients in forest ecosystems. This chapter first presents the methods (critical limits, models and input data) that are needed to calculate critical metal loads (Section 5.2), followed by the results obtained (Section 5.3). This includes critical metal loads in comparison to calculated present loads at 245 Intensive Monitoring plots and in comparison to measured present loads at approximately 20 plots in Germany. Finally, a discussion of the results and conclusions are presented in Section 5.4.

# 5.2 Methods

# 5.2.1 Locations

An overview of available measurements to allow calculations of effect-based critical metal loads (only requires data on the precipitation excess), present bulk metal deposition critical metal loads based on the stand-still approach (requires data on present metal contents in the soil) is given in table 5.1. Data on both bulk deposition were available in Germany for approximately 30 Intensive Monitoring plots. For Cd and Pb, bulk data were also available at nearly 150 plots in Poland, but data on present metal contents, allowing a calculation of critical loads based on the stand-still approach, are not available at those plots. This was only the case at part of the plots in Germany and Austria, but for the latter country, no bulk deposition data are available.

# 5.2.2 Assessing critical loads for heavy metals

## 5.2.2.1 Impacts of heavy metals and critical limits

## **Impacts**

With respect to impacts and risks of heavy metals on terrestrial ecosystems, a major distinction can be made between impacts on humans who use groundwater for drinking water or who consume crops grown on soil (human toxicological risks) and risks to ecosystems itself (ecotoxicological risks). The eco-toxicological risks associated with elevated heavy metal concentrations in terrestrial ecosystems include:

Table 5.1. Number of plots that allow calculations of effect-based critical metal loads ( $CLM_{eb}$ ; plots with data on the precipitation excess), present bulk metal deposition BDM and critical metal loads based on the stand-still approach ( $CLM_{ss}$ ; plots with data on present metal contents in the mineral topsoil) for Cd, Cu, Pb and Zn.

Country	Number	of plots							
	CL <sub>eb</sub>	BDCd	CLCd <sub>ss</sub>	BDCu	CLCu <sub>ss</sub>	BDPb	CLPb <sub>ss</sub>	BDZn	CLZn <sub>ss</sub>
France	25	0	0	0	0	0	0	0	0
Belgium	6	2	0	0	0	2	0	2	0
The Netherlands	1	0	0	0	0	0	0	0	0
Germany	81	31	30	31	31	23	30	40	41
Italy	10	0	0	0	0	0	0	0	0
United Kingdom	10	0	0	0	0	0	0	0	0
Ireland	3	0	0	0	0	1	0	0	0
Denmark	15	0	0	0	0	0	0	0	0
Greece	3	0	0	0	0	0	0	0	0
Portugal	1	0	1	0	1	0	1	0	1
Spain	9	0	0	0	0	0	0	0	0
Luxembourg	1	0	0	0	0	0	0	0	0
Sweden	23	0	0	0	0	0	0	0	0
Austria	17	0	16	0	17	0	17	0	17
Finland	17	0	0	0	0	2	0	7	0
Poland	0	147	0	0	0	148	0	0	0
Slovak Republic	0	0	0	0	0	6	0	0	0
Norway	10	0	0	0	0	0	0	0	0
Hungary	4	0	0	0	0	0	0	0	0
Croatia	1	0	1	0	1	0	1	0	1
Czech Republic	1	0	1	0	1	3	1	3	1
Estonia	4	0	4	0	4	0	0	0	4
Total	242	180	53	31	55	185	50	52	65

- reduced microbial biomass and/or species diversity of soil micro-organisms and macrofungi, affecting microbial processes such as enzyme synthesis, litter decomposition/mineralisation and soil respiration. A review of these effects is given by Bååth (1989).
- a decrease in abundance, diversity and biomass of soil fauna, especially invertebrates such as nematodes and earth worms. A review of these effects is given by Bengtsson and Tranvik (1989).
- reduced development and growth of roots and shoots (toxicity symptoms), elevated concentrations of starch and total sugar and decreased nutrient concentrations in foliar tissues (physiological symptoms) and decreased enzymatic activity (biochemical symptoms) of vascular plants including trees. A review of these effects is given by Balsberg-Påhlsson (1989).
- heavy metal accumulation followed by possible effects to essential organs on terrestrial fauna, such as birds, mammals, or cattle in agricultural soils. Those effects are considered important with respect to Cd, Cu and Hg since these metals can accumulate in the food chain (Jongbloed et al., 1994).

Concern about the atmospheric input of heavy metals (specifically cadmium and lead but also copper and zinc) to terrestrial ecosystems, such as forests, is specifically related to the impact on soil organisms and the occurrence of bio-accumulation in the organic layer (Bringmark and Bringmark, 1995; Bringmark et al., 1998; Palmborg et al., 1998). With respect to copper and zinc, the possible occurrence of deficiencies in view of forest growth is another relevant aspect. Another concern is related to the leaching of metals (specifically cadmium and mercury) to surface water, having an adverse impact on aquatic organisms and causing bio-accumulation in fish, thus violating food quality criteria.

In the past several studies have been carried out to assess critical loads of heavy metals for terrestrial and aquatic ecosystems on national (Bakker, 1995) and European scales (Van den Hout et al., 1999). With respect to terrestrial ecosystems, the attention was focused on forests, where metal deposition is the only external source. The critical load approach can also be applied to agriculture, where the load refers to the input by both fertilisers/animal manure (sometimes also sewage sludge) and atmospheric deposition. In several countries, there is also concern about the excess input of heavy metals (specifically Cd, Cu and Zn) in agriculture (e.g. Moolenaar and Lexmond, 1998). An excess of heavy metals may lead to agricultural products with unacceptable levels of heavy metals and even reduced crop production (Alloway, 1990; Fergusson, 1990). In this case, the critical load approach can give insight into necessary changes in management practices of agricultural land.

# Critical limits

The most commonly available critical limits for heavy metals are critical total concentrations in the soil solid phase. The implicit assumption is that (eco-toxicological) effects are due to metal accumulation in the soil. One of the problems with critical metal concentrations is that the official critical limits for mineral soils are not eco-toxicologically based. This is because background concentrations appear to be higher than maximal permissible concentrations in laboratory toxicity tests. This apparent inconsistency is partly due to differences in metal availability in these toxicity tests and in the field (e.g. Klepper and Van de Meent, 1997). Another reason may be that grown up plants (in contrast to seedlings used in laboratories) have developed defence mechanisms, e.g. through mycorrhizae, and thus neutralise heavy metal toxicity. Considering this inconsistency, it is just as appropriate to use the present metal concentrations as the critical limit for the mineral soil. The critical load then equals the load that does not lead to further accumulation of metals in the soil.

Critical limits for metal concentrations in the soil solution are presently lacking with respect to direct effects on soil organisms. Critical limits based on laboratory studies, using No Observed Effect Concentrations (NOECs) are mostly related to total metal concentrations (Bååth, 1989; Bengtsson and Tranvik, 1989; Witter, 1992; Tyler, 1992). Recently, however, critical limits for dissolved metal concentrations were derived by:

- NOEC toxicity data for soil in view of impacts on plants and microbiota, organisms from which you can be sure that the effect is only through the soil solution
- Data gathered in Germany (Schütze and Throl, 2000), France (Farret and Magaud, pers. comm.) and the Netherlands (Crommentuijn et al., 1997) in which both NOECs and soil properties regulating metal availability (organic matter content, clay content and pH) are available
- Transfer functions for Cd, Cu, Pb and Zn given in Section 5.2.2.2 to calculate related NOECs for soil solution from the soil solid phase and
- Statistical approaches, deriving limits based a log-logistic fit of the NOEC data (Aldenberg and Slob, 1991) and applying a 95% protection level (HC<sub>5</sub>)

Critical limits for metal concentrations in soil solution thus calculated are presented in Table 5.2. More information on the approach, the data sets used and the uncertainties of the derivation are given in de Vries et al. (2002). A separate estimate related to effects on plants is also given. These limits are based on measured Lowest Observed Effect Concentration (LOEC) data from

laboratory studies with culture solutions reported by Balsberg-Påhlsson (1989) applying a safety factor of 10. For Cd, Pb and Zn, these limits are higher but for Cu it is clearly lower.

Table 5.2. Critical limits for dissolved metal concentrations (mg.m<sup>-3</sup>) for Cd, Pb, Cu and Zn based on (i) a statistical interpretation of calculated NOEC data for micro-organisms, soil invertebrates and plants and (ii) measured LOEC data for plants divide by a safety factor of 10

	(ii) meusureu LOE	<i>J</i>				
Metal	Critical limit	Critical limit (mg.m <sup>-3</sup> )				
	NOEC data	LOEC data				
Cd	0.8	2.0				
Cu	16	2.5				
Pb	8	15				
Zn	9	25				

#### 5.2.2.2 Steady-state soil model used to calculate critical loads for heavy metals

#### Calculation of critical loads

As with nitrogen and acidity, critical loads are derived from a steady-state mass balance model. A mass balance including all major metal fluxes (in  $g.ha^{-1}.yr^{-1}$ ) in an ecosystem reads:

$$M_{td} = M_{gu} - M_{we} + M_{er} + M_{ad} + M_{sp} + M_{sr} + M_{bp} + M_{le}$$
(5.1)

where the subscripts td refer to the total deposition gu to root uptake, we to weathering, er to erosion, ad to adsorption, sp to seepage flow, sr to surface runoff, bp to bypass flow and le to leaching. M stands for a heavy metal and can be substituted by the chemical symbol of the individual metal (Cd, Cu, Pb and Zn) under consideration.

As in the previously described critical load models, a steady-state situation is assumed, implying that the critical load is to be valid for an infinitely long period. Adsorption is not included in a steady-state calculation, although adsorption relationships are used to calculate the metal leaching rate when using the stand-still approach (see below). An additional assumption is that the soil is in an oxidised state. The model can thus not be applied to very poorly drained soils with groundwater levels near the surface. The reason is that anaerobic conditions violate the equilibrium partitioning concept due to precipitation of metal sulphides (e.g. Janssen et al., 1996). Note, however, that under such conditions metals are strongly retained (unless a fluctuating ground water level causes acidification) and critical loads are likely to be very high. The limitations of these various assumptions are further discussed in De Vries and Bakker (1998) and De Vries (1999).

In order to apply the model, we further assumed that the soil is a homogeneously mixed soil compartment with only downward transport of water and metals (no seepage flow, surface runoff and bypass flow) is assumed. Furthermore, metal loss by erosion is not included. Using an effect-based approach, based on critical dissolved metal concentrations, erosion should not be included. Erosion removes solid phase material and thus influences the time period before such a steady-state concentration is reached (to be calculated with a dynamic model) but not the dissolved metal concentration itself. That is determined by processes acting on the soil solution, including leaching, uptake and weathering. Using the stand-still approach, it is unclear how erosion should be included. If soil with the same content is eroded as in the layer that we consider (0-10 cm), it does not influence the concentration. We are only left with less soil. Only when the concentration of the eroded material is different, it should be accounted for, but this can theoretically be higher (leads to an increase CL), or lower (leads to a decreased CL). Furthermore, there are not only

places where the soil erodes, but also places where soil accumulates in the landscape. Considering those difficulties, erosion has been neglected both in calculating an effect-based and a stand-still load. Using all those assumptions, Eq. 5.1 can be simplified to:

$$M_{tl} = M_{gu} - M_{we} + M_{le}$$
(5.2)

The critical load, being the acceptable total load of heavy metal inputs by deposition thus corresponds to the sum of tolerable outputs from the system by harvest and leaching minus the natural inputs by weathering release, according to (De Vries et al., 2001):

$$CL(M) = M_{gu} - M_{we} + M_{le(crit)}$$
(5.3)

where:

CL(M) = critical load of heavy metal M (g.ha<sup>-1</sup>.yr<sup>-1</sup>)
 M<sub>gu</sub> = removal of heavy metals by biomass harvesting or net uptake in forest ecosystems, respectively, from the mineral topsoil (g.ha<sup>-1</sup>.yr<sup>-1</sup>)
 M<sub>we</sub> = weathering release of heavy metals in the mineral topsoil (g.ha<sup>-1</sup>.yr<sup>-1</sup>)
 M<sub>le(crit)</sub> = critical leaching of heavy metals from the mineral topsoil (g.ha<sup>-1</sup>.yr<sup>-1</sup>)

The model can be even more simplified by neglecting weathering outside volcanic or ore-rich areas ( $M_w = 0$  in Eq.1). This approach implies that the critical load equals the net uptake by forest growth or agricultural products plus an acceptable metal leaching rate. The considered soil depth for the calculation is 10 cm below the interface of the organic and mineral horizons, which is generally quite homogeneous. Exceptions are some podzolised soils, where this 10 cm soil layer is often not so homogeneous, since the boundary between the A and B horizons is about there. Furthermore, adverse impacts on plants and soil organisms, which are the main target groups considered, is mainly related to this layer (De Vries and Bakker 1998). In deriving critical loads, Eq. (5.3) was used, being in accordance with the countries that recently made such calculation in response to the call for data by the ICP Mapping and Modelling.

## Calculation of the critical metal leaching

The critical metal load can be derived from a critical metal leaching rate, being the product of the percolation flux Q and a critical dissolved metal concentration, according to (compare Eq. 4.5):

$$M_{le(crit)} = 10 \cdot Q_{le} \cdot [M]_{ss(crit)}$$
(5.4)

where:

 $Q_{le}$  = the flux of leaching water leaching from the mineral topsoil (m.yr<sup>-1</sup>)

 $[M]_{ss(crit)}$  = the critical limit for the total concentration of heavy metal in the percolating soil solution (mg.m<sup>-3</sup>)

The value of 10 is needed to convert the unit from  $mg.m^{-2}.yr^{-1}$  to  $g.ha^{-1}.yr^{-1}$ . The total concentration of heavy metal in the soil solution is the most appropriate value to calculate the tolerable leaching flux. In this term both the free metal ions and the metals bound in dissolved complexes are included. Both parts are relevant to the leaching process. In the effect-based approach, values for  $M_{ss}(crit)$  are directly used. In the stand-still approach, however, the present

metal concentration is used as a criterion and the dissolved metal concentration has to be calculated.

Equilibrium processes that determine the partition of metals between various phases are adsorption to the soil and complexation. There are various possible approaches to derive total dissolved metal concentrations (M<sub>ss</sub>) from total soil metal concentrations (M<sub>t</sub>). The simplest approach is a direct empirical approach relating both concentrations, while accounting for the impact of major soil properties influencing the sorption relationship. This approach is, however, not suggested here, because there is no real process mechanism involved in this approach, since part of the metals extracted by aqua regia do not interact with the soil solution (inert part, being equal to the total minus the reactive part). The most fundamental approach is to relate the free metal ion activity to the reactive soil metal content, accounting also for the impact of major ions in soil solution competing with the metals and then calculate the total dissolved metal concentration, M<sub>ss</sub>, from the free metal ion activity, using a (simple) complexation model (De Vries and Bakker, 1998). This approach does, however, require more data and is therefore not suggested yet. In this study, the total dissolved metal concentration, M<sub>ss</sub>, was derived by first calculating the reactive metal concentration, M<sub>re</sub>, from the total metal concentration, M<sub>t</sub> and then deriving the total dissolved metal concentration, M<sub>ss</sub>, from the reactive metal concentration, M<sub>re</sub>, accounting for the impact of soil properties.

The reactive metal concentration (0.43N HNO<sub>3</sub> digestion) was related to the so called total concentration (aqua regia digestion) according to:

$$\log M_{re} = \beta_0 + \beta_1 \cdot \log M_t + \beta_2 \cdot \log(\% OM) + \beta_3 \cdot \log(\% clay)$$
(5.5)

where:

 $M_{re}$  = Reactive concentration of heavy metal M in the soil (mol.kg<sup>-1</sup>)

 $M_t$  = Total concentration of heavy metal M in soil (mol.kg<sup>-1</sup>)

Values for the various coefficients were derived from approximately 630 soil samples in which both the Aqua Regia and 0.43 N HNO<sub>3</sub> extractable metal content was determined together with the soil properties organic matter, clay and pH-KCl. The data originate from (i) a Dutch national inventory on the quality of non-polluted arable soil (312 records), (ii) an inventory on floodplain soils (200 records, both contaminated and non-contaminated soils) and (iii) two smaller studies (49 and 69 records, respectively) on Dutch soils where both samples from the top soil as well as deeper soil layers were included from arable soils and samples taken in natural areas (forest, grassland). Results are shown in Table 5.3. More information on the data set and the optimisation of the parameters is given in Römkens et al. (2002) and de Vries et al. (2002).

 Table 5.3.
 Values for the coefficients  $\beta 0$ - $\beta 3$  in the relationship relating reactive (Mre) and so-called total soil concentrations (Mst) of cadmium and lead, according to Eq. (5.3)

Metal	β <sub>0</sub>	$\beta_1$	$\beta_2$	$\beta_3$	$\mathbf{R}^2$	se-y <sub>est</sub> <sup>1)</sup>
Cd	0.225	1.075	0.006	-0.020	0.82	0.26
Cu	0.400	1.152	0.020	-0.169	0.93	0.13
Pb	0.063	1.042	0.024	-0.122	0.88	0.17
Zn	0.483	1.257	0.198	-0.309	0.96	0.15

<sup>1)</sup> The standard error of the y-estimate on a logarithmic basis

The dissolved total metal concentration was derived from the reactive metal content according to:

$$[M]_{ss} = (M_{re} / K_f)^{1/n}$$

Where

 $K_f$  = Non-linear partition or Freundlich adsorption constant (mol<sup>1-n</sup>.m<sup>3n</sup>.kg<sup>-1</sup>) [M]<sub>ss</sub> = Concentration of metal M in the soil solution (mol.m<sup>-3</sup>) n = Freundlich exponent

To obtain an equation that can be used for a range of soils,  $K_f$  was calculated as a function of the contents of organic matter and clay and pH (thus relating data on those soil properties to the adsorption of metals to the soil solid phase), according to:

$$\log K_{f} = \alpha_{0} + \alpha_{1} \cdot \log(\% OM) + \alpha_{2} \cdot \log(\% clay) + \alpha_{3} \cdot pH$$
(5.7)

The values of  $\alpha_0$ ,  $\alpha_1$ ,  $\alpha_2$  and  $\alpha_3$  were obtained by multiple linear regression. Results of such a fit, based on two data-sets of 114 soil samples and 1466 complete records of both solid phase and solution composition, are given in Table 5.4. More parameters can be included, such as the concentration of dissolved organic carbon (DOC), but those data often lack on larger scale levels. In regressions in which the DOC concentration was included as a predictor variable, the explained variance (R<sup>2</sup>) increased by only 1% for Cd, Pb and Zn and by 3% for Cu. Considering this result and the fact that DOC is only partly available at the Intensive Monitoring plots, this predictor variable was not used in the analyses. More information on the data set and the optimisation of the parameters is given in Römkens et al. (2002) and de Vries et al. (2002).

copper, lead and zinc concentration, according to Eq. (5.5).									
Metal	$\alpha_0$	$\alpha_1$	$\alpha_2$	$\alpha_3$	n	$R^2$	Se-y <sub>est</sub> <sup>1)</sup>		
		(%OM)	(%clay)	(pH)			-		
Cd	-5.01	0.65	0.27	0.29	0.54	0.77	0.37		
Cu	-3.67	0.50	0.18	0.17	0.45	0.63	0.35		
Pb	-3.06	0.85	0.02	0.26	0.67	0.58	0.55		
Zn	-4.96	0.51	0.36	0.52	0.77	0.85	0.41		

Table 5.4Values for  $\alpha o, \alpha 1, \alpha 2$  and  $\alpha 3$ , and n in the transfer function between reactive and dissolved cadmium,copper, lead and zinc concentration, according to Eq. (5.5).

<sup>1)</sup> The standard error of the y-estimate on a logarithmic basis

From Eq. 5.3 and Eq. 5.4 the dissolved metal concentration [M] can be calculated from the total metal concentration in the soil  $ctM_s$ , thus allowing the assessment of a critical metal leaching rate by multiplication with the percolation flux Q.

Taking the minimum critical leaching rate of the criteria related to the present metal concentration in the soil solid phase and the dissolved metal concentration in the soil solution, the critical load will neither cause an accumulation of metals in the soil (stand-still approach) nor affect plants (effect-based approach).

### 5.2.2.3 Assessment of input data

### Metal deposition

Unlike nitrogen and acidity, the present deposition of metals can not simply be calculated by using data on bulk deposition and throughfall, correcting for the effects of element interactions

with the canopy (leaves and needles). The problem is that above-ground foliar uptake of lead is nearly equal to litterfall, since the lead is adhered to the foliar surface and transported to the soil by litterfall. Since litterfall data are not available, the total deposition was simply assumed to equal bulk deposition. This is likely to be an underestimate. A comparison of the Cd, Cu, Pb and Zn content in the organic layer of forested sites and nearby deforested sites in the Netherlands showed ratios of 2.5 for conifers and 1.6 for broadleaves. (De Vries and Bakker, 1998). This might indicate that bulk deposition should be multiplied by forest filtering factors of a similar order of magnitude, but more research on the comparison between total deposition and bulk deposition is needed before such a correction should be applied.

Considering the limited data on bulk deposition, a comparison of present loads with effect-based critical loads was made by not only using measured but also model calculated present deposition data (Smeets et al., 2000). A comparison of present loads with stand-still-based critical loads was made by using measured bulk deposition data only, but this comparison only applies for Germany (see table 5.1).

## Metal weathering

Metal weathering rates for the root zone were derived by multiplying the base cation weathering rates (see Table 4.7) with the molar ratio of the total metal content and the total base cation content in parent material whenever available, as described in De Vries and Bakker (1998). When metal data for parent material were not available, average weathering rates for sandy soils, loamy soils and clay soils were distinguished, based on data given in De Vries and Bakker (1998).

# Metal uptake

As with nitrogen and base cations, net metal uptake was calculated at each site by multiplying the annual average growth rate of stems with the density and the metal content in stems. For the derivation of those data we refer to Section 4.2.3.3, where the derivation of the net uptake of nitrogen is presented. Metal content data in stems (in mg.kg<sup>-1</sup>) were based on De Vries and Bakker (1998) and equalled 5.0 for Pb and Cu, 0.3 for Cd and 25 for Zn. Since critical loads were calculated for the mineral topsoil (0-10 cm), the total net uptake was further multiplied by a root uptake fraction (fraction of fine roots) in this layer and the overlying humus layer, as compared to the total root zone. As a first approximation, this root uptake factor was taken equal to 0.5 (see also Reinds et al., 2001).

## Critical metal leaching

The critical acidity leaching rate was calculated by multiplying the precipitation excess by a critical dissolved metal concentration, either derived from the present metal content, as described above (stand-still principle) or taken directly from the literature (see Table 1; effect-based principle). The precipitation excess was calculated with a water balance model described in De Vries et al. (2001).

# 5.3 Results and discussion

# 5.3.1 Critical loads for metals based on the impacts on soil fauna and plants

## Comparison of average present and critical metal loads

Table 5.5 gives information on average present loads for Cd, Cu and Pb based on model calculations (Smeets et al., 2000) and the average effect-based critical loads for these metals including Zn, based on critical dissolved metal concentrations and a steady-state approach. The table also includes the percentage of plots exceeding critical loads for Cd, Cu and Pb. Critical limits used are for dissolved metal concentrations based on a statistical interpretation of NOEC data in the standard calculation and on LOEC data for plants divide by a safety factor of 10 in an alternative calculation (see Table 5.2). To allow a comparison between present loads and critical loads, results are limited to plots where both data on bulk deposition and metal contents in the mineral topsoil are available (compare also Table 5.1).

 Table 5.5
 Average total present deposition load (PDL), critical load (CL) related to impacts on soil fauna and plants (based on NOEC data) and percentage of plots exceeding the critical load (CL excess) for Cd, Cu, Pb and Zn. Numbers in brackets are based on LOEC data

			s ure bused on LOEC du	u
Metal	Number of PD	L (g.ha <sup>-1</sup> .yr <sup>-1</sup> )	CL (g.ha <sup>-1</sup> .yr <sup>-1</sup> )	CL excess (%)
	sites			
Cd	242	0.4	0.55 (1.3)	29 (6)
Cu	242	3.9	12.5 (2.7)	8 (51)
Pb	242	26	5.8 (10.0)	91 (78)
Zn	242	-	11.2 (20.7)	-

On average the present Pb deposition is much higher than the critical load, whereas the reverse is true for Cu when one uses the critical load based on the NOEC data, while the difference is small for Cd. The number of plots where critical loads were exceeded was 91% for Pb, 29% for Cd and 8% for Cu. When using the critical load based on LOEC data for plants, the number of plots where present loads exceed those critical loads reduce to 78% for Pb and only 6% for Cd, but it increases from 8 to 51% for Cu. This result shows the extreme sensitivity of the results to the value of the (rather uncertain) critical limit. For Zn, no present deposition data were available, but literature data suggest that present loads are generally below critical loads.

Differences between tree species are generally small as illustrated in Table 5.6 for Pb and Cd, but the real effect is probably larger, since the present loads are based on model calculations that do not include the different filtering effects of varying tree species

 Table 5.6
 Average total present deposition load (PDL), critical load based on NOEC data (CL) and percentage of plots exceeding the critical load (CL excess)

Tree	Number of	PDL (g.ha <sup>-1</sup> .yr <sup>-1</sup> )		CL (g.ha <sup>-1</sup> .yr <sup>-1</sup> )		CL excess (%)	
species	sites <sup>1</sup>	Lead	cadmium	lead	cadmium	lead	cadmium
Pine	59	24	0.35	4.3	0.41	87	37
Spruce	96	23	0.32	6.7	0.63	88	17
Oak	30	29	0.47	5.2	0.49	97	40
Beech	44	32	0.47	5.9	0.54	100	41
Other	13	27	0.36	8.1	0.76	92	8
All	242	26	0.38	5.8	0.55	91	29

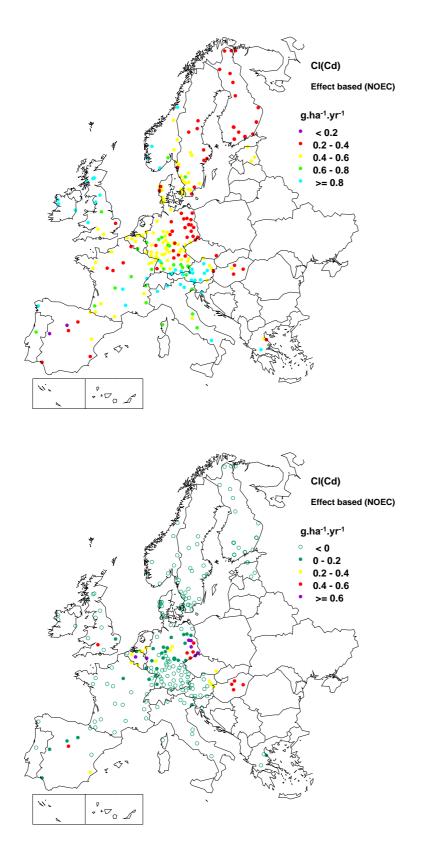


Figure 5.1 Geographical variation of critical loads (top) and critical load exceedances (bottom) for cadmium  $(g.ha^{-1}.yr^{-1})$  related to impacts on soil fauna (acceptable Cd concentration 0.8 g.m<sup>-3</sup>).

### Geographic variation in critical loads and their exceedances

As an example, the geographic variation in critical Cd loads related to impacts on soil fauna (acceptable Cd concentration in soil solution of 0.8 g.m<sup>-3</sup>) and their exceedances by present loads are presented in Fig. 5.1. The results for critical Cd loads (Fig 5.1A) do show a certain north-south gradient, but the pattern is much less distinct than for nitrogen. Unlike nitrogen, the pattern is not so much influenced by tree uptake but by the precipitation surplus. High critical Cd loads (>0.8 g.ha<sup>-1</sup>.yr<sup>-1</sup>) mainly occur in high precipitation areas, such as parts of the UK, Norway and the mountainous areas in Central Europe. It has to be noted that the critical loads are related to a steady-state situation. If one would calculate a target load including an acceptable accumulation in a certain (e.g. 100 year) time period, the pattern would have been much more influenced by differences in soil type. Critical Cd loads are mainly exceeded in plots in Western Europe (Germany, Netherlands, Belgium), where the present loads of Cd are relatively high (Fig. 5.1B).

## 5.3.2 Critical loads for metals and their exceedances based on the impacts on soil

Table 5.7 gives information on average present loads for Cd, Cu, Pb and Zn based on bulk deposition measurements and the average critical loads for these metals, requiring no accumulation of metals in the soil (stand-still approach). The table also includes the percentage of plots exceeding critical loads for these metals. To allow a comparison results are limited to plots where both data on bulk deposition and metal contents in the mineral topsoil are available (compare also Table 5.1).

	percentage of	of plots exceeding the	critical load (CL exces	s) for Cd, Cu, Pb and Zn
Metal	Number of	PDL (g.ha <sup>-1</sup> .yr <sup>-1</sup> )	CL (g.ha <sup>-1</sup> .yr <sup>-1</sup> )	CL excess (%)
	sites			
Cd	23	0.8 (0.20-2.0)	0.20 (0.05 - 0.95)	96
Cu	21	66 (12.2 – 219)	2.8 (0.65 - 7.85)	100
Pb	24	16 (5.0 – 37)	35.7 (2.1 - 367)	46
Zn	21	200 (54 - 967)	217 (54 - 677)	43

 Table 5.7
 Average total present deposition load (PDL), critical load (CL) related to the stand-still approach and percentage of plots exceeding the critical load (CL excess) for Cd, Cu, Pb and Zn

 M total
 Note of plots exceeding the critical load (CL excess) for Cd, Cu, Pb and Zn

For Pb, the critical load at which no further accumulation takes place (stand-still principle) is generally higher than the effect-based critical load. Because these results are based on a very limited number of plots, results should be interpreted with care as they are not representative for all plots in Europe. For example if the average stand-still load for Cd is computed for all plots (n=53) with a measured Cd content (so including also the plots without measured deposition), the value would be 0.75 g.ha<sup>-1</sup>.yr<sup>-1</sup> which is more than 3 times as high as for the 24 plots with deposition. Furthermore, the variation in the bulk deposition of heavy metals is very high (standard deviation is in same order of magnitude as average).

# 5.4 Discussion and conclusions

### 5.4.1 Uncertainties in critical loads

### Limitations of the steady-state model approach

Limitations of the steady-state model approach have already been mentioned in Section 5.1. The approach assumes that the concentration of heavy metals in the soil has reached a steady-state. This assumption signifies that the concentration in the soil does not change in time because the amount of heavy metal entering the considered soil system is equal to the amount that leaves this system. The validity of this assumption depends on the magnitude of the time-scales of the various input and output processes. If for example a heavy metal sorbs very strongly to the soil, it may take hundreds of years before a steady-state is reached, and the concentration in the soil solution has reached a level at which the input by atmospheric deposition or other sources is balanced by leaching and uptake. This must be kept in mind when comparing a present load with the critical metal load. The time period needed to reach steady-state should, in principle, be calculated with a dynamic model, since the rate of accumulation and leaching changes during time. Such models should then also include the possible changes in soil acidity in response to changes in acid atmospheric deposition, in view of the much stronger direct impact of soil acidity changes on metal concentrations than the long term accumulation rates.

### Uncertainties in critical limits, model structure and input data

As with nitrogen and acidity, uncertainties in critical metal loads derived by steady-state soil models are determined by the uncertainty in critical limits, model structure and input data. A systematic overview of these effects is given below.

### Uncertainties in critical limits

Differences in the literature with respect to critical limits for heavy metals are due to differences in (i) effects (species) considered, (ii) laboratory (or field) conditions involved and (iii) extrapolation procedures of single-species toxicity (NOEC) data to a critical value that is assumed to protect an ecosystem sufficiently. The extrapolation procedures are questionable since (i) toxicity data are generally too few to postulate a certain distribution, (ii) it is unlikely that test species are random choices, (iii) laboratory and field conditions, such as pH, clay and organic matter content, influence metal availability and (iv) there are interactive effects (Forbes and Forbes, 1993; Smith et al., 1993). As with nitrogen and acidity, the limits based on the stand-still approach, being the measured present metal concentrations are therefore more reliable than the critical limits based on impacts on either soil organisms or plants. In this study, critical loads in view of soil accumulation could, however, only be calculated for a limited number of plots in Germany.

## Model assumptions

Apart from the assumption of steady state, there are various other assumptions that are used in applying the simple critical load equation (Eq. 5.3), including: (i) the heavy metal present in the soil follows the concept of equilibrium partitioning, (ii) the soil is homogeneously mixed, (iii) the soil is in an oxidised state and there is no seepage flow, (iv) metal cycling can be neglected and (v) adsorption and complexation can be described with relatively simple transfer functions for the

relation between metals in soil and soil solution. More information on those uncertainties is given in De Vries and Bakker (1998). In general one can say that the models used to calculate critical metal loads are not applicable in very wet (anaerobic) circumstances. Other uncertainties, e.g. due to neglecting the metal cycle, are probably less important than the uncertainties due to (unknown) variations in the adsorption constant (see below).

# Input data

The influence of input data depend on the effect considered (critical load in view of soil accumulation or in view of impacts on the soil solution). In general, the uncertainty in the partition (adsorption) constant is the cause of largest uncertainties in the calculated critical loads for terrestrial ecosystems (De Vries and Bakker, 1998). The uncertainty of a steady-state critical load based on a dissolved critical metal concentration is mainly due to the uncertainty in the precipitation excess, apart from the uncertainty in the limit itself.

# **5.4.2** Conclusions

Considering the limitations in the critical load approach, the following preliminary conclusions can be drawn:

- On average the modelled present Pb deposition is much higher than the critical load. When related to the impact on soil fauna (with rather stringent and uncertain limits) exceedances were found on 91% of the plots. Critical load calculated from critical limits for plants were exceeded at 78% of the plots.
- For Cd the difference between modelled deposition and critical load is small, with exceedance on 29% of the plots. For Cu, the critical load is only exceeded in 8% of the plots. Critical loads for plants show an inverse result, with exceedances in only 6% of the plots for Cd, but in 51% of the plots for Cu. This shows the extreme sensitivity of the results tot the value of the uncertain critical limit.
- High critical metal loads mainly occur in high precipitation areas, such as parts of the UK, Norway and the mountainous areas in Central Europe. It has to be noted that the critical loads are related to a steady-state situation. Critical Cd loads are mainly exceeded in plots in Western and Central Europe, where the present loads of metals are relatively high.

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# ANNEXES

### Annex 1 Country results on ground vegetation

#### Background

In December 2001, a questionnaire was send out by the chairman of the Biodiversity Working Group (Pat Neville), in close collaboration with the chairman of the Expert Panel on Ground Vegetation (Dan Aamlid) asking for a summary of any national evaluations carried out on the ground vegetation dataset. The following questions were asked:

- 1. Have any national/regional evaluations been carried out on the ground vegetation data set of the Level II plots ?
- 2. Has any evaluation of the ground flora been carried out at the Level I plots ?
- 3. If not, are there plans to evaluate this data in the future at Level I or Level II? If yes,
- 4. What were the principal sampling methods ?
- 5. What were the principal evaluation methods?
- 6. At how many plots did the evaluation occur ?
- 7. What were the major results ?
- 8. Were any trends in the data apparent ?
- 9. Were any cause-effect relationships detected?
- 10. Is a list of these reports available?

As stated in the request, the purpose of these questions was to collate an overview of national work on the ground vegetation data for the Technical Report 2002. This Annex presents such an overview. First an overview of the approaches is given in Table A.1.1. Results obtained from national evaluations are described below followed by the relevant literature.

#### Overview of results obtained from the national evaluations

#### Austria

Level II: No correlation between species diversity indices of vascular plants (shrub and herb layer) and moss layer (bryophytes, lichens); among all indices of stand structure strong correlations exist; only weak correlations between species diversity indices and indices of stand structure, i.e. only Shannon index of vascular plants showed a negative correlation with Cox index of clumping (and also with standard deviation of DBH) at the 0.05 level. The diversity indices of ground vegetation show significant differences between both applied assessment methods: Plot size and the scale for cover estimation are influencing the results. With the Braun-Blanquet assessment methods diversity indices are higher. My interpretation: Assessing coverage by cover classes overestimates evenness. Level I: Relations were found among Ellenberg's indicator values, as well as between these indicator values and some environmental factors and even species richness: a) A positive relation was found between moisture value and nitrogen.

#### Finland

We have made a general evaluation of the 1998 survey data consisting of all Finnish Level II observation plots (27 on mineral soil and 4 on peatland). This evaluation has been published in the Finnish national report 1999 (Salemaa and Korpela, 2000). It includes tables on the numbers and mean coverages of the plant species in species groups, evenness E and Shannon H' biodiversity indices as well as the Detrended Correspondence Analysis (DCA) ordination in order to find the main compositional gradient of the vegetation data.

France	Finland			Bulgaria		Austria	Country
Jean François Dobremez	Maija Salemaa			Ekaterina Pavlova		Franz Starlinger	Contact Name
Yes	Yes			Yes		Yes	National Evaluation
yes	Yes			Yes		Yes	Level I
yes	Yes			Yes		Level II yes	Future evaluation plans
The principal sampling method was transect method (Braun- Blanquet relevé) on 8 transects (50 x 2 meters) in each plot. 4	Level II: 16 x 2m2 plots		Level I : 0.1-0.2ha plot	Level II: 1x1m plots	Level II: 1 x 225 or 400m2 plot and 10 x 4m2 plots Level I methods not defined	Braun-Blanquet	Country Contact National Level I Future Sampling Methods E Name Evaluation evaluation plans
Statistical and numerical analyses	Generalised Linear Models (GLIM)		Level I : 0.1-0.2ha plot Level I: determination of life plant forms according to Raunkier (1934) and relative species abundance expressed in % using the parameter "projection cover	Level II: Braun-Blanquet	Level II: Indices of species diversity (Shannon, Simpson, evenness), several indices of stand structure; correlation analysis (Spear man's rho); regression analysis Level I: Ellenberg's indicator values; regression analysis.		Evaluation Methods
101	Level II: 31		Level I: 20 plots monitored every 3 years	Level II: 3 plots:	Level II: 20 Level I : 513 s		No of Plots
After 2 campaigns of see summary yes relevés no general trend is detected except the effect of wild big game in relation with fencing.	refer to summary	Trends have been outlined during regeneration and successions. <sup>2)</sup>	Level I: 20 Ecotypical and plots phytocyclic monitored fluctuations in every 3 vegetation have been years established <sup>1)</sup>		None apparent		Data Trends
see summary	refer to summary		none apparent		None apparent		Cause- Effects?
yes	yes		yes		Yes		Reports Available

Russia	Norway	Netherlands	Italy	Ireland	Greece	Germany		Country
Natalia Goltsova	Nicholas Clarke	Han Van Dobben	Roberto Canullo	Pat Neville	George Baloutsos	Ina-Maria Schulze		Contact Name
only for epiphtyic lichens	Yes	Yes	yes	No	no	yes		National Evaluation
only for epiphtyic lichens	Yes	No	no	No	no	yes for Saxony and Baden- Württember g		Level I
yes	Level II: Yes	Level I no Level II yes	Further evaluation are planned combining population data of tree <sup>5)</sup>	Not immediately	yes	yes		Future evaluation plans
Finnish standard SFS- 5670, 1990	Level II: Yes Level II: 50 x 1x1m plots	Braun-Blanquet 1 x 400m2 plot	2 approaches <sup>6</sup> )	Braun-Blanquet 5 x 25m2 plots	n/a	In general 3 different approaches <sup>4)</sup>	relevés inside fence, 4 relevés outside fence	Sampling Methods
n/a	Multivariate statistics, canonical discriminate analysis 12 plots	Multivariate statistics	LII data evaluation was integrated in the frame of National Evaluation. <sup>7), 8)</sup>	n/a	n/a	Species lists, Ellenberg Numbers, sociological classes, species number, diversity indices, ordination methods		Evaluation Methods
150	Level II: s 12 plots	200 19 ICP Forest 181 National	19	9	n/a	, 125		No of Plots
none apparent	No relationship has been found between changes in vegetation composition and a pollution gradient	none apparent	none apparent yet	n/a	n/a	See summary	3)	Data Trends
none apparent	Б	none apparent	see future plans	n/a	n/a	Future work Yes		Cause- Effects?
Yes	yes	Yes	yes	Yes	not yet	Yes		Reports Available

<ol> <li>The quan prevailing si 2) The resul</li> <li>There is <i>z</i> Fagus has in</li> <li>Category B Category C</li> <li>Layers cc</li> <li>a comparent</li> <li>a comparent</li> <li>b coverag</li> <li>50x50 cm ss</li> <li>NFC attee cov</li> </ol>	United Kingdom		Switzerland	Slovakia	Country
<ol> <li>The quantitative correlation between species could be r prevailing species. Regional ranges of variation in the con 2) The results have shown considerable variability in the of 3) There is a lot of browsing on many plants. Covering of <i>Fagus</i> has increased too. Another noticeable temporary ch 4) Category A "Braun-Blanquet-design" plot size 400 m2 Category B large scale sampling unit", size of a sampling Category C "transect-design", size of a sampling unit 1 or 5) Layers collected by " tree growth group", and trying to 2004). Relations among Cryptogamic and Vascular divers 6) - a) coverage estimation (by layer and species), using 6 the fenced plot; analogous spring data when relevant phen in a comparable buffer zone , outside the fenced plot - b) coverage specific estimation, epigeal individuals coun 50x50 cm sampling units; within fenced plot; only on sur 7) The aim was to describe the position of each Plot in a " relative cover data (within plot) and Fisher's alpha index ( 8) NFC attempted also to define the vegetation typology the source of the sample also to define the vegetation typology the source of the sample also to define the vegetation typology the source of the sample also to define the vegetation typology the source of the sample also to define the vegetation typology the source of the sample also to define the vegetation typology the source of the sample also to define the vegetation typology the source of the tempted also to define the vegetation typology the source of the tempted also to define the vegetation typology the source of the tempted also to define the vegetation typology the source of the tempted also to define the vegetation typology the source of the tempted also to define the vegetation typology the source of the tempted also to define the vegetation typology the tempted also to define the vegetation typology the tempted source of tempted source of tempted sour</li></ol>	Andy Moffat No		Anne Thimonier	Pavel Pavlenda	Contact Name
tion between al ranges of v considerable g on many pl unother notice nquet-design" pling unit", s growth groo be growth groo by growth groo typtogamic ar (by layer and (by layer and spring data w spring data w the position, epigea within fenced within fenced plot) and Fisl lefine the veg	ıt No		Preliminary evaluations	No	National Evaluation
species could ariation in the variability in ants. Coverin able tempora 'plot size 400 ize of a samp ize of a samp	No		Preliminary evaluations	No	Level I
be related to t content of ma g of <i>Rubus fru</i> g of <i>Rubus fru</i> g of <i>Rubus fru</i> ry changes are ma ing unit about 1 or 4 m2, siz g to evaluate r g to evaluate r g to evaluate r g to evaluate B ra ng 6 scale B ra phenological s phenological s phenological s counts, wood 1 summer; 3-ye a a "multidime dex (populatio	Not immediately	Level II yes	Level I yes	Yes	Future evaluation plans
<ol> <li>The quantitative correlation between species could be related to the changes in the meteorological and hydrologic prevailing species. Regional ranges of variation in the content of macro- and microelements in the monitoring specie 2) The results have shown considerable variability in the content of microelements as referred to different vegetative 3) There is a lot of browsing on many plants. Covering of <i>Rubus fruticosus</i> have increased sometimes fantastically in <i>Fagus</i> has increased too. Another noticeable temporary changes are due to clearing and to disturbance of soil by wild 4) Category A "Braun-Blanquet-design" plot size 400 m2</li> <li>Category C "transect-design", size of a sampling unit 1 or 4 m2, size of the sampling area about 400 m2</li> <li>Layers collected by " tree growth group", and trying to evaluate relations with changes of the system (deposition, 2004). Relations among Cryptogamic and Vascular diversity and spatial structure of ground Veg. are also planned (s 6) - a) coverage estimation (by layer and species), using 6 scale Braun-Blanquet, in 12 10x10 m alternate sampling un the fenced plot; analogous spring data when relevant phenological spring can be depicted; 3-years records in all 19 (in a comparable buffer zone , outside the fenced plot in a "multidimensional space" defined by sets of variables. Our relative cover data (within plot) and Fisher's alpha index (population level: relation among species richness and individues to describe the position of each Plot in a "multidimensional space" defined by sets of variables. Our relative cover data (within plot) and Fisher's alpha index (population level: relation among species richness and individues to describe the vegetation typology through community within-plots data, using the hierarchica 8) NFC attempted also to define the vegetation typology through community within-plots data, using the hierarchica 8) with the relation typology through community within-plots data, using the hierarchica</li> </ol>	n/a	Level II: 16 1m <sup>2</sup> units and 1 or 2 500m <sup>2</sup> plots	Level I Braun-Blanquet (30 m <sup>2</sup> , 200m <sup>2</sup> , 500m <sup>2</sup> )	n/a	Sampling Methods
/drological reg ng species hav egetative orga stically inside sil by wild boa tit 400 m2 tt 400 m2 tt 400 m2 tt 400 m2 nall 19 (up to n all 19 (up to n all 19 (up to n all 19 (up to n all individua and individua erarchical sint	n/a		Level I Braun-Blanquet Multiple stepwise regression; (30 m <sup>2</sup> , 200m <sup>2</sup> , 500m <sup>2</sup> ) (Canonical) correspondence	n/a	Evaluation Methods
e during the ilso been est of the monit fence and the These two re These two re These two re These two re These two re in a gr i) plots and y i) plots and y i) plots and y related pararected plots ected plots ected plots ected plots ected plots ected plots	n/a	16 Level II	680 Level n/a I	n/a	No of Plots
gime during the years as well as the changes in the life cycle of the re also been established ns of the monitoring species or during the years the fence and the number of seedlings, specially for <i>Abies</i> and rs. These two result in an increase of plant diversity cover/herb population behaviour, etc.) within single plots (say: 003). cover/herb population behaviour, etc.) within single plots (say: 003). arranged in a grid (as a chess table taking up only one colour) within 26) plots and yearly in 10 selected plots; the same in 12 10x10 plot nd related parameters on systematic-adaptive grid system of 100 selected plots ation furnished Shannon-Weiner H' index for community level 1 density). haxonomy of phytosociological approach.	n/a		n/a	n/a	Data Trends
hanges in the g the years s, specially fo plant diversit ) within single ) within single holds; the same olots; the same olots adaptive grid s	n/a		n/a	n/a	Cause- Effects?
ife cycle of the r <i>Abies</i> and y plots (say: ne colour) within in 12 10x10 plot ystem of 100 munity level	n/a		Yes	n/a	Reports Available

The 1998 data was reordinated using the Global Nonmetric Multidimensional Scaling (GNMDS) and fitted environmental vectors (stand variables and chemical composition of the organic layer) for a poster presented in the Heerenveen NFC/SAG meeting in September 2001 (Salemaa et al., 2001). The main compositional gradient in the ordinations represented the change in site fertility, combined with the variation in soil moisture and location along the south-north axis. N and S deposition patterns follow the south-north gradient.

The above-mentioned book (Reinikainen et al., 2000) consists of maps, graphs and tables (abundances and frequencies on the years 1951 - 53, 1985 - 86 and 1995) about 100 most common forest and peatland plant species of Finland. The book has been written in Finnish but it has English summary with a key for maps, tables and figures. The abundance maps are based on kriging interpolation of the plot-wise abundance estimates. Examples of the maps have been presented in the Heerenveen poster (Tomppo et al., 2001). There are some publications on the vegetation on mineral soil (Tonteri et al., 1990a, 1990b; Tonteri, 1994; Oksanen and Tonteri, 1995) and on peat land forests (Korpela and Reinikainen, 1996a, 1996b; Korpela, 1999), which are based on the NFI (Level I) data set. The temporal change in the abundance of common forest floor mosses during 1951 – 95 has been analysed (Mäkipää et al., 2000). In this publication the possible effects of acidification and nitrogen deposition on the relative abundance of *Hylocomium splendens, Dicranum polysetum* and *Pleurozium schreberi* have been discussed. (There is also a submitted manuscript on this topic; Mäkipää and Heikkinen, 2002).

#### France

Evaluations lead to following results:

- plant diversity of plots according to dominant species or according to biogeography.
- The most diversified forests are *Larix decidua* dominated forests and then, *Quercus robur*, *Abies alba, Picea abies, Fagus sylvatica, Pseudotsuga menziesii, Quercus petraea, Pinus sylvestris, Pinus laricio, Pinus pinaster*, mixed *Quercus robur* + *petraea*.
- Most diversified forests occur in mountain biogeographical region, then in Atlantic and then in continental.
- effect of fence on ground vegetation
- achievement of several sampling methods
- variability amongst experts

#### Germany

Evaluations lead to following results:

- The assessment methods differ considerably between the plots and for that reason the comparability of data is influenced. Harmonisation should be aimed concerning sampling design, layer definitions and scales for cover degree (Schulze and Bolte, 2001).
- Aiming at comparable data of biodiversity, species area relationship from each level II-plot should be performed with harmonised methods (Schulze and Bolte, 2001; Seidling, 2001).
- Diversity indices (Shannon-Wiener-index, Evenness) parameterise different properties of the floristic composition of the vegetation cover (Seidling, 2001)
- The vegetation development is influenced to a large extent by climatic conditions, air pollution, game, forest management, development of the stand characteristics and internal processes of vegetation (Seidling, 2001).
- Appropriate evaluation methods concerning the influence of air pollution are the Ellenberg values and different ordination methods (Seidling, 2001)
- Mosses and lichens contribute considerably to biodiversity. They are sensitive bio-indicators due to their anatomical and physiological peculiarities, (Stetzka and Stapper, 2001). Special

attention requires the recolonisation of sensitive species as an effect of reduced  $SO_2$ -input in forest ecosystems and the nitrogen affected spreading of species (Stapper, 2000). These species should be more considered in future.

- Harmonisation of assessment methods and scoring-systems concerning epiphytes in forests is needed (John and Schröck, 2001; Stetzka and Stapper, 2001).

#### Italy

The use of Fisher's alpha diversity index is sensitive enough and can guarantee a relative independence from the dimension of sampled area and can be used for the whole of phytocoenosis as will be combined with Tree Growth data; the dynamical status of the undergrowth can be the major responsible of variation between and within the forest types we sampled; as expected by the sampling design, the vegetation types analysis confirms that there are no replicates of similar situations throughout Italy, so that a lot of forest types are represented (16) by the National LII network allowing to further simple descriptions of forest diversity, at local scale, and as a check-system of eventual generalised models (inferred from phytosociological national data: the Environment Ministry has committed a national evaluation of biodiversity status which uses, for plants, that approach).

#### Norway

In Norway, no relationship has been found between changes in vegetation composition and a pollution gradient. There were small between-years differences and, naturally, greater differences between plots.

#### Netherlands

In the 200 stands of the national network (this includes the 19 ICP-F stands) soil chemistry has been determined in 1995 and vegetation relevees have been made in 1996 (Van Dobben et al., 1997). These data have been used to determine the relationship between soil and vegetation. Additionally, non-soilchemical variables have been used that partly originate from national datasets and models (meteo, deposition). There appears to be a strong relationship between the vegetation and these explanatory variables. The following factors have the strongest influence on the vegetation: light, soil chemistry (especially availability of base cations), atmospheric deposition, and groundwater level. Out of the atmospheric variables, deposition of Mg and SO<sub>x</sub> have a significant effect. Indicators for N deposition (NO<sub>x</sub> and NH<sub>y</sub>) have no significant effect on the vegetation. Out of the soil nutrients, P has a larger effect than N, which indicates N saturation. Also, indications were found for a toxic effect of Al (Van Dobben and De Vries, 2001).

#### Switzerland

The predominant gradients in Swiss forest vegetation are nutrient availability and moisture in terms of primary factors. In terms of secondary factors, the forest vegetation is best explained by the factors degree days, annual rainfall and soil skeleton (Wohlgemuth et al., 2000). The analysis is based on 15'000 available relevés in Switzerland. They are not identical with the level I or level II vegetation data.

#### Literature/ Country Reports

#### Austria

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Starlinger F. and Neumann M. 2001: Quantifizierung von Bestandesstruktur und Diversität. FBVA-Berichte, 120: 59-73

Neumann M., Schnabel G., Gärtner M., Starlinger F., Fürst A., Mutsch F., Englisch M., Smidt S., Jandl R., Gartner K. 2001: Waldzustandsmonitoring in Österreich. Ergebnisse der Intensivbeobachtungsflächen (Level II) FBVA-Berichte, 122: 1-235

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# Annex 2 Analysis of the effect of using bulk deposition, throughfall or total deposition on the relationship with ground vegetation composition.

A selection was made of the plots used for the analysis in the main text for which throughfall and total deposition data were available for all ions. There were 194 plots that met this criterion. It was attempted to improve the homogeneity of this dataset by removing rare species, using the same criteria as in the main text. However, this did not lead to a much higher homogeneity according to these criteria (Table A.2.1) and therefore the data used in this analysis are just a subset of the original data.

action	number of	number of	$GL_1$	$GL_2$	$(\lambda_1 + \lambda_2) / \Sigma \lambda * 100\%$
	species	plots			
dataset used in TechRep	396	360	17.4	36.6	15.7%
select plots with total dep and throughf. data <sup>1</sup> )	382	194	28.3	7.6	13.5%
remove species occurring only once	348	194	28.3	7.6	13.6%
remove species occurring only twice	294	194	19.8	7.6	13.3%

Table A.2.1 Manipulation of species data for additional analysis

1) this dataset was used for analysis

Histograms of the deposition variables were drawn after LN[Y-MEAN(Y)+1] transformation. The distributions tended to be slightly right-skewed and therefore the addition term was increased from 1 to 2.72 (i.e. after transformation the minimum value is ca. 1). This resulted in approximately normal distributions for all deposition variables (incl. bulk deposition) which were used in the analysis. Correlation matrices were made of the deposition variables (Table A2.2). Bulk, throughfall and total deposition are strongly correlated for all ions ( $r \approx 0.8 - 0.9$ ) except K in throughfall (r[bulk]  $\approx 0.5$ ). It can be concluded that the 'additional' deposition variables do not add much information to the predictor set used in the main text except for K in throughfall.

					bulk d	leposition	l		
		NH4	NO3	SO4	Ca	Mg	Κ	Na	Cl
	NH4	0.79	0.74	0.70	0.50	0.34	0.36	0.46	0.48
	NO3	0.72	0.78	0.68	0.49	0.30	0.35	0.40	0.41
II	SO4	0.71	0.72	0.81	0.46	0.40	0.33	0.56	0.61
ghfa	Ca	0.72	0.69	0.76	0.86	0.59	0.56	0.51	0.54
throughfall	Mg	0.54	0.47	0.64	0.63	0.83	0.53	0.77	0.78
thı	Κ	0.68	0.68	0.75	0.62	0.41	0.48	0.54	0.57
	Na	0.41	0.38	0.57	0.26	0.60	0.25	0.91	0.88
	Cl	0.49	0.47	0.65	0.35	0.60	0.28	0.90	0.91
	NH4	0.83	0.73	0.73	0.61	0.45	0.49	0.52	0.53
-	NO3	0.79	0.87	0.79	0.59	0.35	0.42	0.46	0.47
tion	SO4	0.69	0.71	0.81	0.43	0.39	0.30	0.56	0.61
deposition	Ca	0.66	0.61	0.72	0.89	0.60	0.51	0.50	0.54
qel	Mg	0.40	0.31	0.52	0.50	0.90	0.46	0.75	0.76
total	Κ	0.57	0.44	0.59	0.54	0.61	0.84	0.53	0.53
ţ	Na	0.39	0.37	0.56	0.24	0.58	0.22	0.90	0.87
	Cl	0.47	0.45	0.64	0.32	0.58	0.25	0.89	0.90

 Table A2.2
 correlation between bulk deposition and throughfall and total deposition; correlation coefficients for each ion in throughfall and total deposition, respectively, vs.bulk are given in bold.

First the analysis of the main text was repeated with the subset of the data as described above. The result is shown in Table A.2.3 (2nd col.).

	(= main text Table 3.14)			511, 194 piots
predictor set	percentage explained variance	number of predictors	percentage explained variance	number of predictors
all predictors	32%	64	46%	62
only countries	13%	20	17%	19
only environmental variables	24%	44	35%	43
uniquely due to environmental variables	19%		29%	
uniquely due to countries	7%		11%	
undetermined	5%		6%	
full model (countries as covariables)	19%	44 <sup>1</sup> )	29%	43 <sup>2</sup> )
significant model	14%	24 <sup>1</sup> )	19%	18 <sup>2</sup> )
restricted model	10%	12 <sup>1</sup> )	13%	10 <sup>2</sup> )

Table A.2.3 Comparison of main text models (Table 3.14) and the same models for a selection of 194 plotsbulk deposition, 360 plotsbulk deposition, 194 plots

<sup>1</sup>) plus 20 dummy covariables to account for the effect of the countries

<sup>2</sup>) plus 19 dummy covariables to account for the effect of the countries

Compared to the original analysis (Table A.2.3, 1st col.) the total percentage explained variance of the full model strongly increases from 32 to 46%, however the increase in percentage explained variance of the 'significant' and 'restricted' models is much less (from 14 to 19% and from 10 to 13%, respectively). The increase in percentage explained variance is therefore probably for the larger part due to the lower number of plots (and, consequently, the higher number of predictors relative to the number of plots). Also the percentage variance explained by the countries increases from 13 to 17%, although there is one country less (Poland). Next the analysis was repeated with bulk deposition replaced by total deposition, by throughfall, and for a model containing all deposition terms (Table A.2.4).

	total dep	oosition	throug	ghfall			
predictor set	percentage explained variance	number of predictors	percentage explained variance	number of predictors	percentage explained variance	number of predictors	
all predictors	46%	62	46%	62	52%	78	
only countries	17%	19	17%	19	17%	19	
only environmental variables	35%	43	35%	43	42%	59	
uniquely due to environmental variables	29%		29%		35%		
uniquely due to countries	10%		11%		10%		
undetermined	7%		6%		7%		
full model (countries as covariables)	29%	43	29%	43	35%	59	
significant model	19%	18	21%	21	22%	23	
restricted model	13%	10	14%	10	14%	11	

Table A.2.4Comparison of models containing terms for all non-deposition variables plus total deposition (1st col.),<br/>throughfall (2nd col.), or bulk deposition + total deposition + throughfall (3rd col.), for a selection of 194 plots

The result of the forward selection of bulk deposition or total deposition (which hardly differed) is given in Table A.2.5. The resulting significant and restricted models do not contain any deposition term.

		Variable	Р	F	% variance explained	% variance explained (cumulative)
		pH_org	0.001	5.65	2.65%	2.65%
		pine	0.001	3.21	1.41%	4.06%
		oak	0.006	3.07	1.41%	5.47%
	ode	beech	0.001	3.19	1.41%	6.88%
	ď'm	continental	0.011	2.89	1.23%	8.11%
	cte	atlantic south	0.026	2.77	1.23%	9.35%
el	restricted' model	N/C_min	0.001	2.73	1.23%	10.58%
pou	'ré	temperature	0.008	2.42	0.97%	11.55%
lt'n		N/C_org	0.006	2.16	0.97%	12.52%
'significant' model		Ca_org	0.005	2.06	0.88%	13.41%
gnif		pH_min	0.017	2.07	0.79%	14.20%
SI		mountain south	0.035	1.78	0.79%	14.99%
		south	0.042	1.65	0.71%	15.70%
		boreal north	0.094	1.65	0.62%	16.32%
		spruce	0.071	1.5	0.62%	16.93%
		calc soil	0.093	1.47	0.62%	17.55%
		P/C_org	0.108	1.41	0.62%	18.17%
		precipitation	0.061	1.42	0.53%	18.70%
		south high altitude	0.153	1.32	0.53%	19.23%
		K_org	0.124	1.32	0.62%	19.84%
		Na_dep	0.107	1.27	0.44%	20.29%
		(further terms not gi	ven)			

Table A.2.4 Forward selection of variables in CCA, selection of 194 plots, on the basis of a full model containing terms for all non-deposition variables plus bulk deposition or total deposition. Explanation of symbols as in Table 3.15.

The forward selection (Tables A.2.6 and A.2.7) shows that for models containing throughfall, K is selected as the one but most important term (after pH). The biplot for the model containing all deposition terms (Figure A.2.1) shows that a high deposition of K in throughfall is correlated with 'rich forest' species, and with the predictors for these species (high pH, beech, high N/C). Apparently the external recycling (i.e. below-ground uptake followed by above-ground excretion) of K is stronger in forests on rich soils. This is in agreement with existing knowledge. The ordering of the species in the biplot is very similar to the one in Figure 3.15 of the main text, the first axis representing the rich vs. poor gradient, and the second axis the lowland vs. upland gradient.

A summary of the various models that have been tested is given in Table A.2.8, showing that including throughfall in the model leads to an increase in the percentage explained variance. This increase is partly due to K, which is strongly correlated to 'rich' forests, and partly due to the significant effects of anthropogenic deposition terms ( $NH_4$  and  $NO_3$ ). The fact that both terms do not have a significant effect in bulk and total deposition is most likely due to the lower fit of the model when K in throughfall is not included. The impact of N deposition terms now become visible and consequently, the model using throughfall seems most appropriate. Also the proportion of the percentage variance explained by the anthropogenic terms in the total

percentage explained variance increases from ca. 3% to c. 10% when including throughfall in the model (cf. Table A.2.8).

		Variable	P <sub>999</sub>	P <sub>99999</sub>	F	% variance explained	% variance explained (cumulative)
		pH_org	0.001	0.000	5.65	2.65%	2.65%
		K_trf	0.002	0.001	3.42	1.50%	4.15%
		beech	0.001	0.000	3.06	1.41%	5.56%
		oak	0.003	0.001	3.18	1.41%	6.97%
		continental	0.010	0.017	2.92	1.32%	8.29%
	[e]	N/C_min	0.001	0.000	2.8	1.15%	9.44%
	nod	atlantic south	0.020	0.020	2.86	1.23%	10.67%
	ed' 1	temperature	0.014	0.010	2.45	1.06%	11.73%
	rict	pH_min	0.006	0.009	2.2	0.97%	12.70%
	'restricted' model	Ca_org	0.009	0.012	1.89	0.79%	13.49%
	-	N/C_org	0.012	0.019	1.9	0.79%	14.29%
		pine	0.015	0.018	1.86	0.79%	15.08%
		mountain south	0.026	0.031	1.79	0.71%	15.79%
		south	0.036	0.040	1.65	0.71%	16.49%
		calc soil	0.089	0.098	1.5	0.62%	17.11%
		precipitation	0.047	0.047	1.5	0.62%	17.73%
labe		Na_trf	0.047	0.045	1.62	0.62%	18.35%
ũ		P/C_org	0.089	0.099	1.43	0.62%	18.96%
ant		spruce	0.095	0.097	1.4	0.53%	19.49%
nific		NH4_trf	0.088	0.087	1.41	0.62%	20.11%
significant' model		NO3_trf	0.064	0.075	1.48	0.62%	20.73%
		south high altitude	0.162	0.158	1.31	0.53%	21.26%
		K_org	0.136	0.127	1.31	0.53%	21.79%
		Bsat_min (further terms not gi	0.120 ven)	0.124	1.31	0.53%	22.31%

 Table A.2.6 Forward selection of variables in CCA, selection of 194 plots, on the basis of a full model containing terms for all non-deposition variables plus throughfall. Explanation of symbols as in Table 3.15; P999 = P after 999 permutations.

 Pafter 999 permutations.

However, the significance of these anthropogenic terms is always low ( $P \approx 0.08$ ) and according to the traditional criterion P<0.05 they would be considered non-significant. In the present analysis this criterion has been relaxed because of the intrinsic uncertainty of the P values determined by bootstrapping (terms have been included in the 'significant' models until P>0.1 for all remaining terms). To estimate the uncertainty in the P-values, these were also determined on the basis of 9999 instead of 999 samples for the model containing throughfall terms. However, the P<sub>9999</sub> values were only slightly different from the P<sub>999</sub> values (Table A.2.6), and therefore the P values given have to be considered reliable.

		Variable	P	F	% variance explained	% variance explained (cumulative)
		pH_org	0.001	5.65	2.65%	2.65%
		K_trf	0.002	3.42	1.50%	4.15%
		beech	0.001	3.06	1.41%	5.56%
	lel	oak	0.003	3.18	1.41%	6.97%
	'restriced' model	continental	0.01	2.92	1.32%	8.29%
	ed'	Na_tot	0.001	2.85	1.23%	9.53%
	stric	atlantic south	0.028	2.61	1.06%	10.58%
	'res	precipitation	0.001	2.43	1.06%	11.64%
		pH_min	0.005	2.31	0.97%	12.61%
labo		N/C_min	0.007	2.13	0.88%	13.49%
шC		Ca_org	0.01	1.95	0.88%	14.38%
'significant' model		N/C_org	0.025	1.93	0.79%	15.17%
nific		pine	0.029	1.67	0.71%	15.88%
sign		mountain south	0.049	1.60	0.62%	16.49%
		calc soil	0.079	1.59	0.71%	17.20%
		south	0.058	1.51	0.62%	17.82%
		temperature	0.078	1.42	0.53%	18.35%
		P/C_org	0.092	1.42	0.62%	18.96%
		spruce	0.101	1.39	0.53%	19.49%
		NH4_trf	0.08	1.43	0.62%	20.11%
		NO3_blk	0.079	1.49	0.53%	20.64%
		NO3_trf	0.082	1.42	0.62%	21.26%
		NH4_tot	0.092	1.35	0.53%	21.79%
		Bsat_min	0.105	1.33	0.53%	22.31%
		K_org	0.111	1.33	0.53%	22.84%
		south high altitude	0.161	1.27	0.53%	23.37%
		(further terms not gi	ven)			

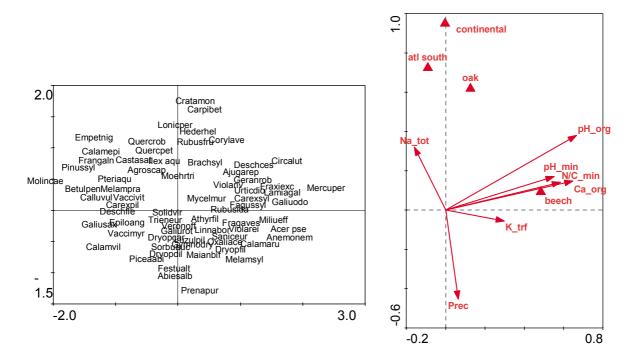
 Table A.2.7 Forward selection of variables in CCA, selection of 194 plots, on the basis of a full model containing terms for all non-deposition and all deposition variables. Explanation of symbols as in Table 3.15

Table A.2.8 Summary of effect of variables in the 'significant' models using various selections of plots and deposition variables. Figures are percentages explained variance.

Variable group	360 plots, bulk deposition (= main text Table 3.16)	194 plots, bulk or 194 plots,194 plots,total depositionthroughfalldeposition		
Actual soil situation	5.8%	7.8%	7.6%	7.5%
Climate <sup>1</sup>	4.9%	6.1%	5.6%	5.2%
Tree species	3.1%	4.9%	4.1%	4.1%
Deposition: non-anthropogenic (K, Na)	0.3%	0.0%	2.1%	2.7%
Deposition: anthropogenic (NH <sub>4</sub> , NO <sub>3</sub> )	0.4%	0.0%	1.2%	2.3%
SUM	14.5%	18.7%	20.7%	21.8%

<sup>1</sup> Includes climate zone, altitude, temperature, precipitation

The main conclusion of this extra analysis is that including throughfall may make it easier to show an effect of anthropogenic deposition. Although in the present analysis the effect of the anthropogenic variables is only weakly significant, they would probably have been significant at P<0.05 if observations from more plots had been available. It is therefore recommended to take throughfall samples in all plots where bulk deposition is also sampled.



*Figure A2.1* Biplot of species and predictor variables in the restricted model including all deposition variables, first and second axis. Explanation as in Figure 3.15 in main text.

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