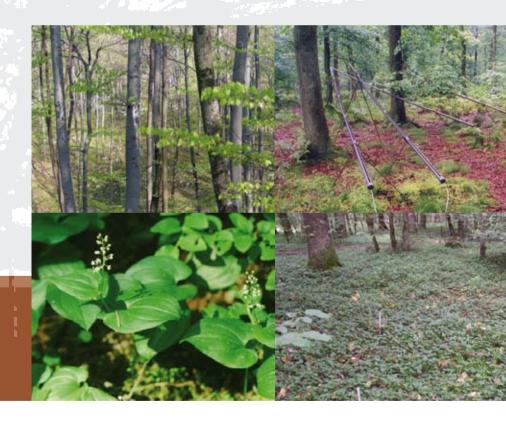


# What happened to our forests in the last decades?

Results of more than ten years of forest ecosystem monitoring in the Netherlands

E.E.J.M. Leeters W. de Vries T. Hoogland B. van Delft R. Wieggers D.J. Brus A.F.M. Olsthoorn H. van Dobben A. Bleeker



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agriculture, nature and food quality



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ABSTRACT

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More than 10 years of forest monitoring in the Netherlands have shown a significant worsening of the crown condition of Douglas fir and Oak over the period 1990 – 2006. This worsening is not related to the atmospheric input, since there is a significant decrease in SO<sub>4</sub> deposition at all plots and a downward trend in N deposition at part of the plots. Deposition of base cations shows a slight but significant increase. The deposition changes are correlated with a significant decrease in the N-content and S-content in the needles of Douglas-fir and Pine over the period 1990-2005. For Oak, the correlations are not significant. The Mg-content increases significantly for all tree species, the K-content in Douglas-fir only, whereas changes in P- and Ca-content are not significant. Finally, there is a significant decrease in SO<sub>4</sub> concentrations decrease significantly at part of the plots. The results indicate a decrease in acidification, related to a decrease in SO<sub>4</sub> and Al concentration, but the problems with N deposition remain, specifically unbalanced nutrient ratios in the foliage, due to the ongoing high N deposition, even though the trend is downward.

Keywords: forests, monitoring, acidification, atmospheric deposition, species composition, element budgets

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# Preface

As a result of forest decline, being originally ascribed mainly to air pollution, the United Nations Economic Commission for Europe (UN/ECE) in 1985 established the International Co-operative Programme on the Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests), which was joined by the EU. The main objectives of this monitoring programme are to: (i) provide a periodic overview on the spatial and temporal variation in forest condition on a European scale by a large-scale systematic network (16 x 16 km; level 1 plots) and (ii) contribute to a better understanding of the relationships between the condition of forest ecosystems and anthropogenic and natural stress factors through intensive monitoring on a number of selected permanent observation plots (level 2 plots).

In the Netherlands, 14 level 1 plots were installed in 1987, with continuous forest vitality data since then. Since 1997, there are only 11 plots left. In 1995, 14 level 2 plots were selected and installed, and the following inventories were made: (a) Crown condition: interval 1 year since 1984, (b) Foliar chemistry: interval 2 years since 1990, (c) Deposition: interval 1 year since 1995, (d) Soil chemistry: in 1990 and 1995, (e) Soil solution chemistry: interval 1 year, since 1990, (f) Increment: interval 5 years, since 1995, (g) Vegetation: interval 5 years since 2000. The reason for having data before 1995 is because these plots were part of a previous the national Forest Health Monitoring Network.

This report presents the trends in crown condition at both level I and level II plots since 1984. Furthermore, we present trends in atmospheric deposition since 1995 and in foliar chemistry and soil solution chemistry since 1990. Finally we present changes in forest increment for the period 1995 -2005, in species composition of ground vegetation since 2000 and in element budgets for the period 2003-2005.

In short, the results indicate a significant worsening of the crown condition of Douglas fir and Oak since 1990, but this worsening is not related to the atmospheric input, since there is a significant decrease in SO<sub>4</sub> deposition at all Level II plots and a downward trend in N deposition at part of the plots. The deposition changes are correlated with a significant decrease in the N-content and S-content in the needles of Douglas-fir and Pine. There is a also significant decrease in SO<sub>4</sub> concentrations in soil solution at all plots since 1990-2001, whereas NO<sub>3</sub>, Al and Mg concentrations decrease significantly at part of the plots. The results indicate a decrease in acidification, related to a decrease in SO<sub>4</sub> and Al concentration, but the problems with N deposition remain, specifically unbalanced nutrient ratios in the foliage, due to the ongoing high N deposition, even though the trend is downward.

We thankfully acknowledge all the colleagues who assisted in carrying out the various surveys. We furthermore thank J.C.H. Voogd for data processing. Finally we thank the European Commission DG Agriculture and the Dutch Ministry of Agriculture nature and Food Quality for funding this monitoring work.

# Summary

#### Background, aim and set up of the Dutch monitoring programme

From the late 1970s onwards, the condition of tree crowns was observed to deteriorate in several forest areas of Europe. As a result of this decline being originally ascribed mainly to air pollution, the United Nations Economic Commission for Europe (UN/ECE) in 1985 established the International Cooperative Programme on the Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests). On 17 November 2003, Regulation (EC) No 2152/2003 concerning monitoring of forests and environmental interactions in the Community (Forest Focus) was implemented, related to the period 1 January 2003 until 31 December 2006. The main objectives of the UN/ECE and EU monitoring programme are to: (i) provide a periodic overview on the spatial and temporal variation in forest condition on a European scale by a large-scale systematic network (16 x 16 km; the so called level 1 plots) and (ii) contribute to a better understanding of the relationships between the condition of forest ecosystems and anthropogenic (in particular air pollution) as well as natural stress factors through intensive monitoring on a number of selected permanent observation plots spread over Europe (the so called level 2 plots).

In the Netherlands, 14 level 1 plots were installed in 1987, with continuous forest vitality data since then. Since 1997, there are only 11 plots left. Tree species in the plots are representative for the Dutch forest on sandy soils. Soil chemistry was assessed in 1995 for the humus layer and the mineral soil layers 0-10 and 10-20 cm below surface. The large scale survey was extended by an intensive monitoring programme of forest ecosystems by installing 14 level 2 plots in 1995. The plots were selected based on a division over the main tree species, deposition level and chemical composition of the soil solution. These level 2 plots are also part of the national Forest Health Monitoring Network. Inventories made on the level 2 plots are: (a) Crown condition: interval 1 year since 1984, (b) Foliar chemistry: interval 2 years since 1990, (c) Deposition: interval 1 year since 1995, (d) Soil chemistry: in 1990 and 1995, (e) Soil solution chemistry: interval 1 years since 2000. An overview of methods used for the collection of data in each survey is given in detail in the main report.

#### Trends in atmospheric deposition of nitrogen and sulphur between 1995 and 2005

Plots of the temporal variation in total deposition, as derived from throughfall and bulk deposition data, for the sites Hardenberg, Zeist, Dwingeloo and Speuld show a declining trend for the deposition of  $SO_4$  in the period 1995-2005, most clearly noticeable from 2000 onwards. Deposition levels of total N, potential acid and base cations show more erratic variation over the years 1995-2005.

Trend analysis shows that the deposition of  $SO_4$  decreases significantly, with 64 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> on average, while the decrease is about twice as high for Dwingeloo

(102 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>). Deposition of total N shows a less clear but still significant decrease of 61 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. The deposition of total-N split up into  $NH_4$  and  $NO_3$  shows a significant decrease for both of respectively 47 and 14 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>. Estimates of trends in nitrogen compounds for individual locations show a significant decrease only for Zeist (total N,  $NH_4$  and  $NO_3$ ) and Dwingeloo (total N and  $NO_3$ ). Deposition levels of base cations for all locations shows a slight but insignificant increase.

#### Trends in crown condition between 1990 and 2006

Plots of the crown condition, in terms of a change vitality class, over the period 1990 - 2006 (data before 1990 were not considered reliable) showed the following trends:

- For Scots pine, there is a recovery at all level I plots, but at level 2 plots there has been a decline around 1993 and there are no signs of recovery until 2006.
- For Douglas fir the forest condition gets worse around 1993 and there are no signs of recovery until 2006.
- The condition for Oak becomes steadily worse over the years.

Trend analysis shows that defoliation of Oak increases significant; 0.77% per year on average. There is also a large and significant overall increase in defoliation of Douglas fir (1.24%/yr),, whereas no significant change was observed for Pine. There is no apparent relation between the declining trends in deposition level and the generally slightly decreasing trends in vitality. Vitality is the reflection of many influences and it is not clear on this moment what causes the low vitality and its erratic temporal variation.

#### Trends in the chemical composition of foliage between 1990 and 2005

Plots of the chemical composition of the foliage show that the N contents of all tree species are almost always above a threshold. This indicates an increased risk for stress induced by drought, frost and diseases. However, the N concentrations show a declining trend and near 2005, they reach the threshold. Inversely, the S contents are always below an upper threshold and show an even stronger declining trend. The other nutrients (P, K, Ca and Mg) are never in the deficiency range, although the Ca and Mg contents in the needles of Scots pine are near the low threshold. The fluctuations of these nutrients in time are fairly large, with no clear visible trend. Results show a sometimes unbalanced nutrition due to high nitrogen contents, specifically with respect to P for Douglas-fir (19% of the plots), Ca for Oak (46% of the plots) and both Ca and Mg for Scots pine (25-30% of the plots).

Trend analyses show that the N-content and S-content in the foliage decrease significantly over the period 1990-2005 for Douglas-fir and Pine but not for Oak. The Mg-content in the foliage increases significantly in the same period for all tree species. The K-content increases significantly in needles of Douglas-fir only, whereas changes in P-content and Ca-content are not significant.

#### Trends in the chemical composition of the soil solution between 1990 and 2001

On average, the concentrations of all major ions in the soil solution, including  $SO_4$ ,  $NO_3$ ,  $NH_4$ , Al, Ca, Mg and K, are comparable in the topsoil and subsoil. In both the

topsoil and subsoil, the  $SO_4$  concentrations show a significant overall decrease for all the tree species over the period 1990-2001. The highest decrease in  $SO_4$ concentrations occurs under Douglas and the lowest decrease under Oak. For NH<sub>4</sub>, there is only a significant decrease in the topsoil for all tree species, being highest under Douglas fir and lowest under Oak. The NO<sub>3</sub> concentration only decreases significantly under Pine, specifically in the subsoil. Furthermore, there is a significant decrease in Al and Mg concentrations in the subsoil for all tree species, except for Al under Oak.

#### Relations between trends in atmospheric deposition and forest ecosystem responses

To analyze if a relation between deposition, foliar chemistry and chemistry of the soil solution exists cross correlation tables were made. Most striking correlations were found between the deposition and foliar content of S, Mg and K for Oak and Pine (R>0.9 except for K in pine with R=0.65), while much lower correlations were found for Douglas (R near 0.3 for S and Mg and absent for K). Pine also shows strong correlations for N in deposition and foliage (R =0.89). There are no strong correlations between similar nutrients in deposition and soil solution. The highest correlation found is for SO<sub>4</sub> and NH<sub>4</sub> in deposition and the concentration of both nutrients in the subsoil, respectively (R near 0.5). Correlations between nutrient deposition and foliar chemistry appear to be much stronger indicating a more direct link between deposition and foliar chemistry.

#### Changes in the increment of trees in the period 1995-2005

The repeated data on tree diameter (at breast height) and tree height in 1995, 2000 and 2005 were used to calculate standing wood volume and changes therein. The carbon pool changes in stem wood were derived by multiplying tree volume with wood densities and tree carbon contents (assumed at 50%). Largest stem wood volumes were generally found for Douglas-fir, with the exception of one plot (Schoonloo). Except for this plot, the stem volumes of Douglas fir sites are above 250 m<sup>3</sup>.ha<sup>-1</sup> and average growth rates are near 25 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup> in the period 1995-2000. Scots pine plots on average show intermediate stem volumes (176 m<sup>3</sup>.ha<sup>-1</sup>) and growth rates (10 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup>), whereas the Oak plots on average show the smallest stem volumes (160 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup>) and growth rates (8 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup>). The carbon pool changes are generally 4 times as low as the volume changes, being near 6 ton C.ha<sup>-1</sup>.yr<sup>-1</sup> for Douglas fir and near 2.5 ton C.ha<sup>-1</sup>.yr<sup>-1</sup> for both Pine and Oak.

#### Changes in the plant species diversity in the period 2000-2005

The variation in the species composition of the ground vegetation was measured in 2000 at 14 plots and in 2005 at 5 plots only. Before determining the magnitude, direction and possible causes of vegetation changes, the variation in the vegetation at a single point in time (i.e. 14 plots in 2000) was evaluated first. We then evaluated the change between 2000 and 2005. Both univariate and multivariate ordination techniques were used to perform the analyses. On the basis of the analysis of the species composition in 2000, it can be concluded that (i) the plots are poor in species, (ii) the differences in species composition can largely be explained from the difference in dominant tree species (Scots pine, Douglas-fir or Oak) and (iii) the species composition indicates acid and nutrient poor conditions. These conclusions

could be further elaborated with the aid of soil chemical data. On the basis of the analysis of the change in species composition in 2000 and 2005 at 5 plots, it can be concluded that (i) the changes between 2000 and 2005 were small and the small number of plots hardy allowed the detection of a pattern in the change and (ii) no abiotic changes are evident from the vegetation changes. This analysis was, however, strongly limited due to the scarcity of data.

#### Element budgets for nitrogen, sulphur, base cations and aluminium

The fate of nitrogen, sulphur, base cations and aluminium was studied in three plots (Hardenberg, Dwingeloo and Zeist) for the period 2003-2005 by assessing element budgets. Considering the element budgets of Cl and Na, which generally behave as a tracer, there is likely to be nearly complete N retention at Dwingeloo, an intermediate N retention (near 50%) at Hardenberg and hardly any N retention at Zeist. For all other elements we could not derive a budget at Zeist, since the conditions were too dry to measure element concentrations in soil solution. At Hardenberg, there seems to be retention of sulphur and base cations, but also a strong mobilization of Al. The release of Al, which is the clearest signal of soil acidification, is mainly due to the external input of SO<sub>4</sub> and NO<sub>3</sub>. At Dwingeloo, there is hardly any soil acidification due to the retention of S and N in the soil profile.

#### Conclusions

More than 10 years of forest monitoring have shown the following trends

1. A significant worsening of the crown condition of Douglas fir and Oak, in terms of an increase in defoliation and a decrease in vitality class, over the period 1990 - 2006. For Scots pine, there is a recovery at all level I plots, but at level 2 plots there has been a decline around 1993 and there are no signs of recovery until 2006.

2. A significant decrease in  $SO_4$  deposition at all 4 plots (Hardenberg, Zeist, Dwingeloo and Speuld), whereas downward trends in nitrogen compounds are only significant for Zeist and Dwingeloo for the period 1995-2005. There is significant increase in N/S ratio on all plots but no significant trend in the NH<sub>4</sub>/NO<sub>3</sub> ratio.

4. A significant decrease in the N-content and S-content in the needles of Douglas-fir and Pine over the period 1990-2005. For Oak, the changes are not significant. The Mg-content increases significantly for all tree species, the K-content in Douglas-fir only, whereas changes in P- and Ca-content are not significant.

5. A significant decrease in  $SO_4$  concentrations in soil solution in both the topsoil and subsoil for all tree species over the period 1990-2001. Furthermore, there is a significant decrease in Al and Mg concentrations in the subsoil for all tree species, except for Al under Oak. The NO<sub>3</sub> concentration only decreases significantly under Pine, specifically in the subsoil.

There are strong correlations between deposition and the chemical composition of foliage, specifically between the deposition and foliar content of S, Mg and K for Oak and Pine. Correlations between nutrient deposition and soil solution chemistry appear to be much less. The results indicate a decrease in acidification, related to a decrease in  $SO_4$  and Al concentration, but the problems with N deposition remain, specifically unbalanced nutrient ratios in the foliage, due to the ongoing high N deposition, even though the trend is downward.

# 1 Introduction

# 1.1 Background and aims of the UN/ECE and EU monitoring programme

# Background

From the late 1970s onwards, the condition of tree crowns was observed to deteriorate in several forest areas of Europe. As a result of this decline being originally ascribed mainly to air pollution, the United Nations Economic Commission for Europe (UN/ECE) under its Convention on Long-range Transboundary Air Pollution (CLRTAP) in 1985 established the International Cooperative Programme on the Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests).

One year later, in 1986 Member States of the European Union (EU) agreed upon the European Union Scheme on the Protection of Forests against Atmospheric Pollution. The Regulation (EEC) No 3528/86 and Regulation (EEC) No 2158/92 dealt with the protection of forests against atmospheric pollution and fire and formed the legal basis for co-financing the relevant assessments. These regulations expired on 31 December 2002 and were, on 17 November 2003, replaced by Regulation (EC) No 2152/2003 concerning monitoring of forests and environmental interactions in the Community (Forest Focus), related to the period 1 January 2003 until 31 December 2006.

Since 1986 the monitoring of forest condition and its development has been carried out in close co-operation under both programmes of UN/ECE and EU. At present, 38 European countries as well as the United States of America and Canada are participating in the programmes, which include assessments according to harmonised methods. These methods are recorded in the ICP Forests Manual (UN/ECE, 1998). Updates of this manual are published on the ICP Forests site: <u>http://www.icpforests.org</u>. The collective monitoring of forest condition of UN/ECE and EU constitute one of world's largest biomonitoring systems, which pursue the objectives of Resolution S1 of the Strasbourg Resolution, H1 of the Helsinki Resolution and L2 of the Lisbon Ministerial Conference on the protection of forests in Europe. The programmes also have developed as an important platform for the exchange of expert knowledge. The monitoring system and the results achieved by it are not only relevant for clean air policies, but also for other processes of international environmental policies, such as those for biodiversity, sustainable forest management and climate change.

ICP Forests, launched in 1985 by the Executive Body of the CLRTAP and operating under the Working Group on Effects (WGE), is mandated to monitor air pollution effects on forests and to contribute to a better understanding of cause-effect relationships. Participating countries nominate and finance their own National Focal Centres (NFC) who are responsible for the collection, validation, evaluation and storage of their monitoring data and aggregation of national data in accordance with the ICP Forests Manual. The NFCs have the task to submit data and accompanying information to the co-ordinating institutes in accordance with the deadlines agreed and the format laid down in the ICP Forests Manual. They can also evaluate and interpret their national data and participate in the evaluation and interpretation of the data at European level. The NFC of the Netherlands is installed at the Ministry of Agriculture, Nature and Food quality, Directie Kennis Ede.

#### Aims

The main objectives of the UN/ECE and EU monitoring programme are to:

- Provide a periodic overview on the spatial and temporal variation in forest condition as well as natural stress factors on an European scale by a large-scale systematic network (level 1),
- Contribute to a better understanding of the relationships between the condition of forest ecosystems and anthropogenic (in particular air pollution) as well as natural stress factors through intensive monitoring on a number of selected permanent observation plots spread over Europe (level 2) and to study the development of important forest ecosystems in Europe.

The current Regulation establishing Forest Focus seeks to implement forest monitoring and protection activities in the following areas:

- Protection against atmospheric pollution;
- Prevention of fires and their causes and effects;
- Biodiversity, climate change, carbon sequestration, soils and protective functions of forests,
- Continuous evaluation of monitoring activities.

Under this scheme, networks of observation points and plots (system for monitoring effects of air pollution) are to be maintained in order to produce periodic inventories and carry out continuous monitoring of forest ecosystems. The forest fire information system will continue to be developed. To achieve the objectives of Forest Focus, two-year national programmes are to be set up by the Member States. They are to include an ex-ante evaluation. Member States are also to submit midterm and ex-post evaluations.

# 1.2 Background and aims of the Dutch monitoring programme

# Background

Starting 1986 ICP Forests in cooperation with the EU (see Section 1.1) established an extensive systematic large scale network (16 x 16 km) of forest samples point, the so called level 1 plots. The aim of this large scale grid net is to get more accurate knowledge of the extent, dynamics and spatial distribution of the symptoms of forest damage in Europe and establish a database for time series analyses of crown condition. The Netherlands started forest vitality monitoring in 1984. The condition of the Dutch forest was observed nation wide on 1500 (being a subset of 3000) sites each year over the period 1984 – 1994, specifically in view of the potential adverse effect of air pollution. Since 1986, it has submitted data for these Level 1, plots to the programs of UN/ECE and EU.

In 1990, a study was carried out at many forested plots to gain a better understanding of cause-effect relationships and this study was repeated in 1995. Since then, data for a number of these forest stands (the so-called level 2 plots) have also been submitted to the programs of UN/ECE and EU. In the beginning the coordination and data collection was steered by the NFC itself. Starting 2000 most of the data collection and later on also the coordination was conducted by Alterra.

Since the start of the monitoring of forest vitality in the Netherlands, there has been a large decline in atmospheric deposition of acidifying compounds, specifically of sulphur and to a lesser extend of nitrogen. This is illustrated in Figure 1.1. This figure presents the trends in total annual deposition of  $SO_x$ ,  $NO_y$  and  $NH_x$  over the period 1981 – 2004, based on a combination of atmospheric measurements and modelling. In this period the atmospheric deposition of sulphate diminished by almost 80%, comparable to the reduction in the emission of sulphur dioxide, both in the Netherlands and abroad. Over the same period the deposition of nitrogen oxides and ammonia decreased with approximately 25%, which is slightly less than the decrease in emission of approximately 30%. At present, nitrogen is the largest contributor to potential acid deposition.

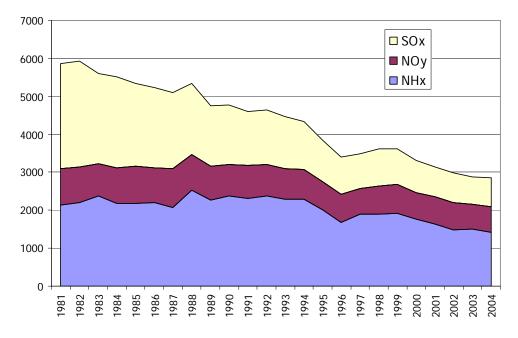


Figure 1.1 Trends in the deposition of  $SO_x$  ( $SO_2$  and  $SO_4^{2-}$ ),  $NO_y$  (NO,  $NO_2$  and  $NO_3^{-}$ ) and  $NH_x$  ( $NH_3$  and  $NH_4^+$ ) in the Netherlands between 1981 and 2004. (Source: De Haan et al., 2007).

#### Aims

A crucial question is in which way the large decline in atmospheric deposition of sulphur and to a lesser extend of nitrogen has affected Dutch forest ecosystems. The main objective of the Dutch national forest monitoring programme is to gain insight in the effects of present emission control measures in view of air pollution and climate change. Forests are monitored in terms of impacts on biodiversity (species diversity of the ground vegetation), forest growth (carbon sequestration) and forest health in relation to nutrient availability and soil and ground water quality. Specific objectives of the Dutch Forest Monitoring Programme are the assessment of:

- Responses of forest ecosystems to changes in air pollution by deriving trends in stress factors and ecosystem condition.
- Critical loads of atmospheric deposition, related to the chemical ecosystem condition, in relation to present loads by evaluating the fate of atmospheric pollutants in the ecosystem in terms of accumulation, release and leaching.
- Criteria and indicators for sustainable forest management, including (i) the role of forests as a net carbon sink to reduce the build up of atmospheric greenhouse gasses, (ii) forest ecosystem health and vitality, (iii) forest production, (iv) species diversity of ground vegetation and (v) protective functions of soil and water resources.

In this context, monitoring is needed of:

- Nutrient availability in trees (foliage) and soils
- Growth and regeneration of forest
- Carbon pools in standing biomass and soil and the changes therein
- Biodiversity in terms of species composition of the ground vegetation.
- Atmospheric deposition and climatic parameters

to determine the impact of air pollution and climate change (external impacts in general) on forests in terms of biodiversity, forest growth and forest vitality.

# 1.3 Aim and contents of this report

The major aims of this report, and the related contents of chapters, are to:

- Give an overview of the results of more than 10 years of forest monitoring in the Netherlands, including an overview of methods used for data collection (Chapter 2).
- Assess trends in atmospheric deposition of nitrogen and sulphur to forest in the Netherlands since 1995 (Chapter 3
- Evaluate the data on trends in time for crown condition (since 1984) and the chemical composition of foliage and soil solution (since 1990) and their possible relations with trends in atmospheric deposition (Chapter 4).
- Assess changes in the increment of trees and in plant species diversity of the forest undergrowth in the period 1995-2005 (Chapter 5).
- Evaluate the fate of atmospheric pollutants in the ecosystem in terms of accumulation, release and leaching by assessing element budgets for nitrogen, sulphur, base cations and aluminium (Chapter 6).

The first part of this report, Chapters 1 and 2, give an overview of the background of the monitoring, historical aspects, the locations, the methods used for assessments and adjustments or calculations carried out to make the data suitable for submission or evaluation. Part of Chapter 2 concerns the national database itself. All data mentioned in this report are stored in an access database which can be installed on your own personal computer or consulted directly from the enclosed CD. For this purpose a users friendly interface is build in the database.

The second part of this report is dedicated to the evaluation of the data. Chapter 3 gives an overview of the total atmospheric depositions over the period 1995 - 2005, based on measurements of bulk deposition and throughfall. It also includes an assessment of the significance of changes in the deposition levels with time. Chapter 4 is dedicated to data on crown condition at 11 level 1 plots and 14 Level 2 plots based on ongoing measurements in the period 1984-2006. It further includes results on the chemical composition of foliage over the period 1990 - 2005 and soil solution over the period 1990 – 2006. The chemical characteristics of foliage are compared to national and international values concerning sufficient or insufficient nutrient availability. Chapter 4 also includes an assessment of the significance of changes in crown condition and in the chemical composition of foliage and soil solution with time. Chapter 5 is dedicated to changes in forest growth and plant species diversity of the ground vegetation over the period 1995 - 2005. In Chapter 6, water fluxes and nutrient budgets are presented for the years 2003-2005. These budgets are based on an assessment of element leaching from measured soil solution chemistry and modelled water fluxes, combined with atmospheric deposition data. These budgets allow the calculation of nitrogen retention and thereby of carbon sequestration. In Chapter 7 finally, conclusions on the changes in our forest over the last decade are given.

# 2 Data assessment

# 2.1 Locations

# Level 1 plots

In the Netherlands, 14 level 1 plots were installed. Table 2.1 gives an overview of the characteristics of these plots and Figure 2.1 shows there distribution over the Dutch forest area. Tree species in the plots are representative for the Dutch forest on sandy soils. Soil chemistry was assessed in 1995 for the humus layer and the mineral soil layers 0-10 and 10-20 cm below surface. Assessments of crown condition are carried out yearly from 1987 till 2006. The assessment of the plots 41 Odoorn, 68 Hellendoorn and 134 Beestenveld were cancelled in 1993, 1996 and 1997 respectively. These level 1 plots are no longer monitored.

Table 2.1 Characteristics of the level 1 monitoring sites

Nr.	location	Tree <sup>1)</sup>	Age <sup>2)</sup>	Soil <sup>3)</sup>	Coordina	ates		
					Х	у	Latitude	Longitude
29	Norg	SP	68	Gleyic Podzol	224200	558900	53°00'50	06°25'15
39	Dwingeloo	SP	79	Haplic Arenosol	226400	539600	52°5020	06°26'45
41	Odoorn	DF		-	250600	541900	52°51'25	06°48'20
59	Ommen	OA	57	Gleyic Podzol	227900	501600	52°29'50	06°27'40
66	Nunspeet	SP	81	Haplic Podzol	186100	487500	52°22'28	05°50'40
68	Hellendoorn	DF		-	225800	492100	52°24'45	06°25'38
76	Sprielde	OA	79	Cambic Podzol	173100	471700	52°14'00	05°39'12
89	Uddel	DF	72	Cambic Podzol	190700	473100	52°14'45	05°54'40
123	Stoppelbergen	SP	73	Haplic Arenosol	082500	376400	51°22'20	04°20'50
125	Ulvenhout	OA	100	Umbric Gleysol	114500	396200	51°33'15	04°48'10
129	St.Anthomis	SP	69	Haplic Podzol	185300	403100	51°37'00	05°49'30
132	de Kempen	OA	68	Gleyic Podzol	144800	372050	51°20'18	05°14'30
134	Beestenveld	DF			183000	395400	51°32'55	05°47'30
136	Leende	SP	70	Gleyic Podzol	164300	373400	51°21'02	05°31'17

1) DF = Douglas-fir; SP = Scots pine, OA = Oak

2) Age calculated for the year 2004

3) Soil type according to the ICP Forests Manual (UN/ECE, 1998)

Level 1 plots are identified by x and y coordinates according to the Dutch *Rijksdriehoekstelsel* and a translation of these coordinates into latitude and longitude. These coordinates make it possible to recover the forest stand. Within this forest stand, trees (circa 20) used for the assessment of the crown condition are marked with yellow paint. There is no further identification of the plot area.

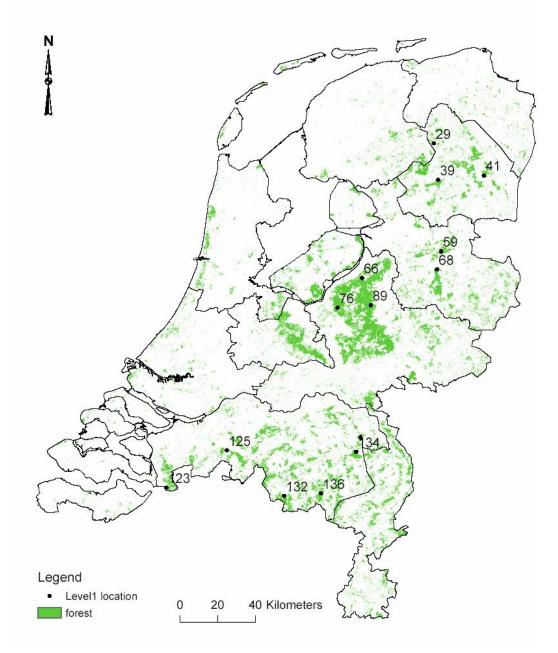


Figure 2.1 Distribution of level 1 plots over the Dutch forest area

# Level 2 plots

In order to contribute to a better understanding of the impact of air pollution and other stress factors which influence the forest ecosystem, the large scale survey was extended by the intensive monitoring programme of forest ecosystems, level 2. The execution of this second level of monitoring was also in close cooperation with ICP forests. Common assessment methods were defined and used. The level 2 plots were installed in 1995. In the Netherlands 14 plots were selected based on a division over the main tree species, deposition level and chemical composition of the soil solution. An overview of the inventories made at the level 2 plots, with their time interval is given in Table 2.2. Monitoring of the level 2 plots started officially in 1995. For some plots however data were sampled also in years before. Crown condition was carried out since 1984 (as part of a larger network of plots that focused on crown condition only), while foliar, soil and soil solution chemistry started in 1990. Table 2.3 gives an overview of the characteristics of the level 2 plots and Figure 2.2 shows there distribution over the Dutch forest area.

Survey	Nr of plots	Measurements since	Interval
Crown condition	14 (5 since 2004)	1984	1 year (since 1984)
Soil chemistry	14 (5 since 2004)	1990	5 year 1990, 1992, 1995
Foliar chemistry	14 (5 since 2004)	1990	(2 year) until 2005
Increment	14 (5 since 2004)	1995	5 yearly: data for 1995, 2000, 2005
Vegetation	14 (5 since 2004)	2000	5 yearly: data for 2000 and 2005
Deposition	4-5	1995	biweekly, yearly calculated values since 1995 to 2005
Soil solution chemistry	14 (5 since 2004)	1990 (1992)-2001	1 year
	3	2003-2005	biweekly; yearly calculated
	2	2006	values since 1995 to 2005

Table 2.2 Overview of available measurements at the Dutch level 2 monitoring plots

Table 2.3 Characteristics of the level 2 monitoring sites

Nr.	Name	Tree <sup>1)</sup>	Age <sup>2)</sup>	Soil <sup>3)</sup>	Groundwater <sup>3)</sup>	Coordin	ates		
			_			Х	у	Latitude	Longitude
39	Gasselte	DF	56	cHd23	VIIId	245950	554900	52°58'22	06°44'28
58	Smilde-1	SP	55	Zn21	VIIId	219000	543750	52°52'49	06°20'36
61	Smilde-2	SP	59	Zn21	VIIId	219850	543950	52°52'49	06°21'16
82	Schoonloo	DF	31	Hn21gF	VIIo	244850	547000	52°54'21	06°43'34
106	Hardenberg	DF	84	Zn21	VId	234000	506870	52°32'42	06°33'00
128	Kootwijk-1	OA	69	Y21	VIIId	178500	465500	52°10'58	05°44'30
129	Kootwijk-2	DF	54	Y21	VIIId	179500	465500	52°10'58	05°45'14
174	Leende-1	SP	65	Hd21F	VIIId	163850	370000	51°19'45	05°31'04
175	Leende-2	SP	69	Zd21F	VIIId	163900	371000	51°19'19	05°31'04
226	Ulvenhout	OA	57	Hn23F	VIIId	111950	395950	51°33'13	04°46'04
1012	Speuld-1	OA	76	gY30gF	VIIId	172000	471850	52°14'08	05°38'13
1040	Zeist	OA	76	zEZ21g	VIIId	143950	457850	52°06'32	05°13'50
2080	Hoenderloo	OA	87	gcY30	VIIId	189050	461900	52°09'88	05°53'10
2084	Speuld-2	DF	42	gY30F	VIIId	175000	473000	52°16'03	05°44'17
2085	Dwingeloo	SP	79	Zd21	VIId	226180	539200	52°50'20	06°26'45
1) DE	- D	cn = c	<b>,</b>	·	0.1				

<sup>1)</sup> DF = Douglas-fir; SP = Scots pine, OA = Oak

<sup>2)</sup> Age calculated for the year 2004

<sup>3)</sup> Soil type according to the Dutch soil classification

<sup>4)</sup> Groundwater class according to the Dutch classification: VIIId means a mean highest groundwater level below 140 cm and a mean lowest groundwater level below 160 cm. For the classes VIId, VIIo and VId these values are respectively 80-140 and >180; 80-140 and 120-180; 40-80 and >180

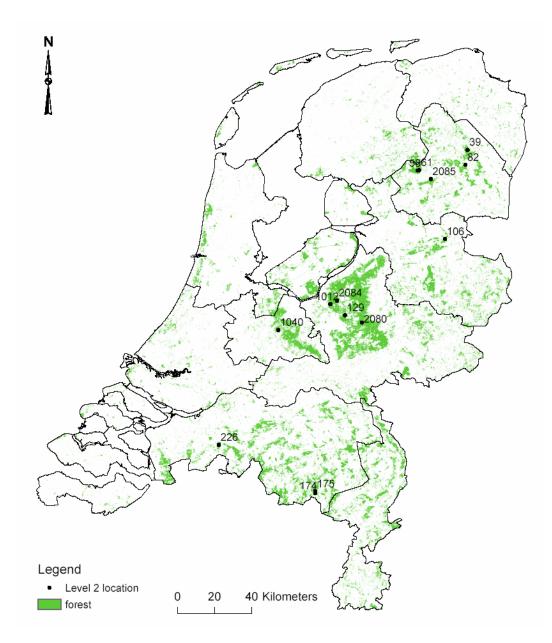


Figure 2.2 Distribution of level 2 plots over the Dutch forest area

Before the start in 1995 of the ICP Forests programme concerning the level 2 plots, 12 of these plots were already monitored over the period 1992 – 1994 in a national programme. The first inventory on these plots was in 1990 in the context of a study concerning the chemical composition of the humus layer, soil and soil solution of 150 forest stands on non-calcareous sandy soils in the Netherlands De Vries and Leeters (2001). In this study foliage and groundwater were also sampled in all 150 forest stands. Furthermore all 150 forest stands were part of the national inventory on forest vitality. This explains why for some parts of the level 2 inventory data are also available before 1995.

In 1994 one plot, 128 Kootwijk-1, was eliminated, since it was no longer accessible for reasons of nature conservation. When in 1995 ICP Forests and the EU started the monitoring of the level 2 plots, three extra plots were added, so there was a total of 14 level 2 plots to start with. In 2002 again a plot was eliminated, 82 Schoonloo. This forest stand was felled completely for the increase of a nearby exploitation of sand mining. In 2005 the total amount of level 2 plots was reduced to 5 (see Table 2.2).

Level 2 plots are also identified by x and y coordinates according to the Dutch Rijksdriehoekstelsel and by latitude and longitude. The outline of the plot area (circa 0.250 ha) is indicated with angles and distances from a starting point which is always located at an intersection of forest paths and indicated by x and y coordinates of the Rijksdriehoekstelsel. Within the plot the location of the trees is determined by their position compared to the halfway line (as an x-as) and the breadth line (as an y-as). This way the plot is divided into two parts an upper (A) and a lower part (B). All trees are numbered according to the following principle. Trees (25) used for the assessment of the crown condition got numbers 1 - 100 (numbers 1 - 25 at the start of the monitoring and the numbers 26 - 100 for replacements of this original 25 trees over the years) Trees in part A got numbers between 101 and 549. Trees in part B got numbers 550 – 999. This information is processed in a GIS and maps are drawn. Figure 2.3 and Figure 2.4 show these maps for plot 106 Hardenberg. This kind of identification is made for all level 2 plots. These maps are all stored in the national database of these monitoring locations (see section 2.3). Prints can be made on A3 format so that the information is better legible. In 1995 when the measurements for the identification of the plot and trees were done, positions of the starting points were drawn on topographical maps from which the x and y coordinates were determined. Using a GIS it seemed that those coordinates were not accurate enough. Therefore small corrections are made.



Scale 1:25000 Coordinates according to Rijksdriehoekstelsel X 23384 Y 506875



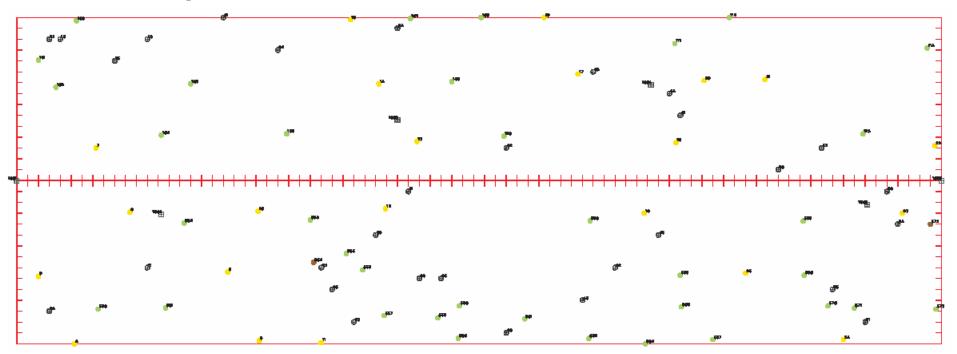
Scale 1:5000 Angle from starting point 335 Distance from starting point 23 m Angle to plot axis 64 Distance to plot axis 36 m Angle of plot axis 64 Distance plot axis 85 m

Figure 2.3 Identification of plot 106 Hardenberg at scale 1: 25000 and at scale 1: 5000

24

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Forest Focus 106 Hardenberg



- ⊞ marker
- tree in plot
- tree in crown assessment
- tree cut
- tree standing dead
- measurement soil solution

Figure 2.4 Identification of plot 106 Hardenberg at scale 1: 212

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# 2.2 Methodological aspects

In this chapter a description is given of the sampling method, sampling periods, chemical analyses and the quality assessments that are made to data in order to make them suitable for submission to either to ICP Forests or the EU programme (see Section 1.1). Attention is given also to the data storage.

# 2.2.1 Deposition

Deposition was first measured in 1995. For the period of October 1995 – October 1996 throughfall was measured at the complete set of 14 level 2 plots. Bulk deposition was measured on a nearby location in the open field for two plots only (106 Hardenberg and 175 Leende-2). In October 1997 the throughfall measurements were continued at four of the 14 plots (106 Hardenberg, 1040 Zeist, 2084 Speuld-2 and 2085 Dwingeloo see Table 2.3). From January 1998 on throughfall measurements are made continuously at the four plots which were also measured at the last quarter of 1997. Bulk deposition measurements are only made at plot 106 Hardenberg. Starting 2003 a fifth plot (175 Leende-2 see Table 2.3) is added. On this plot both throughfall and bulk deposition is measured. All measurements were carried out by the Energy research Centre of the Netherlands (ECN). Table 2.4 gives an overview of the measurements and analyses carried out over the period 1996 – 2005. For all other plots bulk precipitation data were made available by the RIVM from their Air Quality Monitoring Network (Van Elzakker & Buijsman, 1999; Van Elzakker, 2001).

For throughfall measurements at each plot 10 gutters were installed in two parallel lines with 5 collectors each at distances of 1 - 2 m. The gutters are 4 m long and have a collecting area of about 400 cm<sup>2</sup>. They were placed with an angle of 150°, about 1.5 m above the surface. Sample bottles were placed below the surface and CHCl<sub>3</sub> was added as a preservative. The samples were kept in the dark at temperatures between 4 and 8 °C below the soil surface. Sample bottles are collected two-weekly and the gutters are rinsed with demineralised water. Five samples were combined into one sample. Two-weekly samples were combined in the laboratory to obtain monthly samples.

Year	Number of plots	S <sup>1)</sup>	Ana	ılysed	elen	nents												
			Quan	Ηd	Cond	К	Ca	Mg	Na	$N-NH_4$	C	N-NO <sub>3</sub>	$S-SO_4$	Alka	N-tot	M	Mn	Fe
1996	14	1	х	Х	х	х	х	х	х	х	х	х	х	х	х			
1996	2	2	х	х	х	х	х	х	х	х	х	х	х	х	х			
1997	4	1	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
1997	1	2	х	х	x	х	х	х	x	х	х	х	х	х	х	х	х	x
1998	4	1	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
1998	1	2	х	х		х	х	х	х	х	х	х	х					
1999	4	1	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
1999	1	2	х	х	х	х	х	х	х	х	х	х	х					
2000	4	1	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
2000	1	2	х	х	х	х	х	х	х	х	х	х	х					
2001	4	1	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
2001	1	2	х	х	х	х	х	х	х	х	х	х	х					
2002	4	1	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
2002	1	2	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
2003	5	1	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
2003	2	2	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
2004	5	1	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
2004	2	2	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
2005	5	1	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х
2005	2	2	Х	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х

Table 2.4 Number of sampled plots and analysed elements for deposition over the period 1996 - 2004

<sup>1)</sup> S = sample code (1 = throughfall, 2 = bulk)

Bulk deposition was measured at the nearest open field using an open bulk deposition collector. The bulk precipitation gathered by the RIVM was done with a wet-only collector. Wet-only collectors are only open during periods of precipitation. Open collectors therefore receive a certain amount of dry deposition. Table 2.5 shows the coordinates of the bulk deposition measurements.

Plot	Collected by	Coordinates			
		X	у	Latitude	Longitude
106	ECN	233637	507515	+523259	+063248
175	ECN	164046	371114	+511945	+059101
1040	RIVM	140600	456900	+520601	+051037
2084	RIVM	177700	476000	+521618	+054311
2085	RIVM	259100	544300	+525233	+065601

Table 2.5 Bulk deposition measurements and their location

Analyses were all made by the ECN laboratory. Before submitting the data to the EU data checks with respect to (i) the balance between cat ions and anions (ii) the difference between measured and calculated electric conductivity and (iii) the ratio between ion concentrations on an individual and an annual basis were preformed as described in de Vries et al. (1999). Result of these checks and a presentation of the data results are over the years reported in Erisman et al. (1997), Erisman et al. (2001), Erisman et al. (2002), De Groot et al. (2003) and Bleeker et al. (2004).

# 2.2.2 Foliage

The chemical composition of foliage was first measured in 1990. In the period 1992 - 1997 it was measured every year and from 1999 on every second year. Table 2.6 gives an overview of the measurements and analyses carried out in foliage during the period 1990 - 2005. Table 2.6 also shows that over the period 1990 - 1994 there was a set of 12 plots. In 1994 one plot was eliminated, it was no longer accessible for reasons of nature conservation. When in 1995 ICP Forests and the EU started the monitoring of the level 2 plots, three extra plots were added, so there was a total of 14 level 2 plots. In 2003 four oak plots were not measured because in august, when the sampling should take place, it was unclear whether or not the sampling could be financed. One plot, 82 Schoonloo was completely cut down.

In the graphs shown in Figure 4.2 (section 4.1), data are plotted as an average of the tree species Douglas-fir, Scots pine and Oak, usually the average of four or five plots. For these tree species data, also averages, from two Dutch national forest surveys, executed in 1990 and 1995 respectively are included. Averages for Douglas-fir come from 16 and 27 forest stands, for Scotch pine from 43 and 42 forest stands and for Oak from 28 and 51 forest stands respectively. In 2000 again a national forest survey was carried out including the chemical characterisation of the foliage. These data unfortunately behaved like outliers in our graphs, especially for nitrogen in Oak, and for phosphorus in all tree species. To avoid repeated descriptions of unexplained exceptions in the data series we decided to exclude the data of 2000 of this report.

Year	Number of plots	Ana	alysed	l cont	ents											
		Ν	S	Р	Ca	Mg	Κ	Na	Zn	Mn	Fe	Cu	Pb	Al	В	С
1990	12	х	х	х	х	Х	х					x <sup>1)</sup>				
1991	-															
1992	12	х	х	х	х	х	х	х	х			х		х		х
1993	12	х	х	х	х	х	х	х	х			х		х		
1994	11	х	х	х	х	х	х	х	х			х		х		
1995	14	х	х	х	х	х	х	х	х	х	х	х		х		
1996	14	х		х	х	x	х	Х	х			х		х		
1997	14	х	х	х	х	х	х	x <sup>2)</sup>	х		х	х		х		
1998	-															
1999	14	х	х	х	х	х	х		х	х	х	х		х		
2000	-															
2001	14	х	х	х	х	х	х		х	х	х	х		х		
2002	-															
2003	9	х	х	х	х	х	х		х	х	х	х		х		
2004																
2005	9	х	х	х	х	x	х		х	х	х	х		х		

Table 2.6 Number of sampled plots and analysed contents of foliage over the period 1990 - 2005

<sup>1)</sup> In 1990 Cu was only analysed for four plots (39, 82, 106 and 129)

<sup>2)</sup> In 1997 Na was only analysed for four plots (82, 226, 2084 and 2085)

Foliage is collected and analysed according to the guidelines by the Committee for Advice on Forest Fertilization (LNV, 1990) and the guidelines as described in Van

den Burg and Schaap (1995). Both guidelines are in line with those of the ICP Forests Manual, Part IV sampling and analyses of needles and leaves (UN/ECE, 1998) and its updates.

Plots with Oak were always sampled in August and plots with Douglas-fir and Scots pine in the period October - November. In these months, the nutrient levels are fairly constant, and can be compared to standards for deficiency and excess. The number of sampled trees depends on the development and height of the plot. In plots with a height less than 20 meters, ten trees were sampled. In plots with a height of 20 meters or more, five trees were sampled. The sampled trees belong to the predominant or dominant classes. Only leaves and needles of the current year were taken. For analyses composite samples were made by mixing equal quantities of each of the five or ten samples. Sampling usually was done by the same team.

Analyses for the years 1995 and 1997 were performed by the Institute for Forestry and Nature Management (IBN). Analyses since 1990 up to 2001 were carried out by the Bedrijfslaboratorium voor grond- en gewasonderzoek (Blgg) in Oosterbeek. Analyses for the years 2003 and 2005 were carried out by the Chemical and Biological Soil Laboratory of Wageningen University, using the same methods as used in the earlier analysis (IBN and BLGG).

European laboratory comparison tests were participated 1995, 1997, 1999 and 2001. In 1995 and 1997 no systematic errors were found (Bartels, 1996, 1998). In 1999 and 2001 the results were also satisfactorily (Bartels, 2000, 2002). In 1999 sometimes with slightly above average values (nearly always < 10% deviation). In 2001, the results were even better, with slightly below average values, all within the critical zone. This deviation from the average is also dependent on laboratories with an anomaly for certain nutrients, as they are included in the average.

# 2.2.3 Soil

Soil samples on level 1 plots were taken only once in 1995. Soil samples on 14 level 2 plots were taken in the years 1990, 1992 and 1995. In 1990 the sampling of the soil was part of a study concerning the chemical composition of the humus layer, mineral soil and soil solution of 150 forest stands in the Netherlands, De Vries and Leeters (2001). The sampling methods for soil and soil solution were the same. For a description of the solid phase and another part for analysing the soil solution using a centrifuge method to extract soil solution. In 1992 soil samples were taken in the framework of a national monitoring programme. The sampling method was the same as in 1990. In 1995 the ICP Forests programme on monitoring level 2 plots began, and the sampling methods changed. For a description of the changes see also Section 2.2.4. These soil samples were all taken by Alterra (or formerly the DLO Winand Staring Centre). Analyses of pH, pH-KCl, Organic Matter, total contents of C, N, P and S, pools of readily dissolvable Al, Fe and P were carried out by the Bedrijfslaboratorium voor grond- en gewasonderzoek (Blgg) in Oosterbeek. Al other

analyses were carried out by the Alterra laboratory. Table 2.7 gives an overview of the sampled layers and analysed elements in soil during the period 1990 - 1995. In 1992 not only the chemical elements were analysed but also the granular composition was analysed. This was also done by the Blgg. Since 1995, no further soil sampling has taken place at the level 2 plots.

# 2.2.4 Soil solution

Soil solution measurements must be divided into two periods. In the first period 1990 – 2001 soil solution was sampled by taking soil samples and using a centrifuge method to extract soil solution. This destructive way of sampling took place only once a year. This method was used only by a minority of member states and was no longer accepted by the EU because the results can not be used for calculations of input – output budgets of chemical component in the soil. Therefore the method was changed in 2002. From 2002 on forwards the soil solution is sampled by means of tension lysimeters with a frequency of 12 times a year. Both methods are described below. Data were only available for the last three months of 2002. Consequently, the results of the measurements from 2003 onwards were used to calculated input – output budgets of the nutrients as described in Chapter 6.

#### First period 1990 – 2001

#### Sampling period

During this period soil samples were usually taken in the months April or May because in this time of the year the element concentrations in the soil solution approached best the annual flux weighted concentrations. Some years however, because of pragmatically reasons, sampling could not (or not completely, as in 2000) be done in this period and was delayed until October, the second best time of the year. Some years also only 7 of the 14 plots were sampled (as in October 2001). In this case measurements always took place on the plots where deposition is measured.

#### Sampling method

In 1990 the sampling of the soil solution was part of a study concerning the chemical composition of the humus layer, mineral soil and soil solution of 150 forest stands in the Netherlands. (De Vries & Leeters, 2001). At each plot, composite samples were taken, consisting of 10 sub-samples for the humus layer and 20 sub-samples for the mineral soil at the depths of 0 - 30 cm and 60 - 100 cm. The number of sub-samples (20) was based on the results from a comparative study of three methods to extract soil solution. The special pattern in which the samples were taken was based on a square in the middle of the forest stand. In this square the samples were taken at approximately 5 m apart and in the middle of the square a description of the soil profile was made.

Year	Layer	Pa	ıram	eter	s an	alys	ed																														
		Hq	pH-KCl	CaCO <sub>3</sub>	OM	C tot	N tot		S tot	Al ox	Fe ox	P ox	H ex	Al ex	Fe ex	Mn ex	Ca ex	Mg ex	Na ex	K ex	NH4 ex	CEC	Al tot	Fe tot	Ti tot	Ca tot	Mg tot	Na tot	K tot	Mn tot	Si tot	Cr tot	Cu tot		Zn tot	Cd tot	Pb tot
Level 1																																					
1995	LFH				х		х	х														х	х	х		х	х	х	х	х		х	х	х	х		
	0-10				х		х	х														х	х	х		х	х	х	х	х		х	х	х	х		
	10-20				х		х	х														х	х	х		х	х	х	х	х		х	х	х	х		
Level 2																																					
1990	LF	х	х		х	х	х	х	х				х	х	х	х	х	х	х	х	х	х	х	х	х	x	х	x	х	х	х	х	х	х	х	х	х
	Н	х	х					х																						х							
	LFH	х	х		х	х	х	х	х																					х							
	0-30	х	х	х	х	х	х	х		х	х	х	х	х	х	х	х	х	х	х	х	х															
1992	LFH	x	X		x	x	x	х	x				x	х	x	x	x	x	x	x	x	x	x	x		х	x	x	x			х	x	x	x	x	x
	0-10	X								x	x																			х	x						
	10-30	x																												x							
	30-60	X																												x							
	60-	x																												x							
	100																																				
1995	LF	х	х		х	х	х	х	х				х	х	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х		х	х	х	х	х	х
	Н		х					х						х														х						х			
	LFH		х					х						х														х						х			
	0-10		х					х						х																				х			
	10-30		х					х						х																				х			

Table 2.7 Layers and analysed soil parameters over the period 1990 - 1995

This way of sampling was also continued in the years 1992, 1993 and 1994. The sampling depths however were changed to 0 - 10 cm, 10 - 30 cm, 30 - 60 cm and 60 - 100 cm. Starting 1994, solution was also centrifuged from the humus layer.

In 1995 when the ICP Forests and EU programs on monitoring level 2 plots began, the spatial pattern in which the samples were taken changed and the number of subsamples taken for the mineral soil became now 25. The sample points in the plot were chosen according to a steady pattern related to (25) trees marked for the assessment of the crown condition. These trees are gathered in groups of five, scattered through the forest stand. In most forest stands these groups lie in a straight line. In the middle of the groups a marker is placed 30 cm below surface. This marker can be tracked with a detector. The line connecting the middles of the groups forms the baseline. Sub-samples were taken on certain points at a distance of 5 m apart in both sides of the baseline. Marked trees and markers are shown in Figure 2.4. In most cases the group of the first 5 trees is not in the plot, the others are. For more information see also Leeters and De Vries (2001) Annex 1.

The humus layer was sampled with a cylinder of steel with a diameter of 14.8 cm. In 1990 and 1995 for every sub-sample of the humus layer the thickness of the litter (L), fermented (F) and humus (H) horizon plus the total thickness was noted. In these years the humus layer was sampled separately as L&F and H horizons when the thickness of the H horizon was 1 cm or more. Otherwise just one humus layer sample was taken. Mineral soil samples were taken with a gutter. With the exception of 1990 and 1995 the sampling was done by the same person

#### Analysed elements

Table 2.8 gives a overview of the number of sampled plots and the analysed elements over the period 1990 – 2001. The most important pool of nutrients in forest is the humus layer. Consequently, the contents of all major nutrients, i.e. C, N, P, K, Ca, Mg and S were measured for this layer for the level 2 plots. In the Level 1 plots, it was limited to organic matter, N and P (Table 2.8). Contents of C (and/or organic matter), N and P were also measured for the mineral layer since the C/N ratio is likely to increase due to the high N input whereas P is an important nutrient whose availability might be limited due to acidification.

In order to gain insight in the buffer characteristics of the soil, the exchangeable cation contents (H, Al, Fe, Ca, Mg, K, Na and  $NH_4$ ) and the CEC were measured both in the humus layer and the mineral topsoil in the level 2 plots. In the mineral layer the pools of readily dissolvable Al and Fe hydroxides were measured in 1990 and 1992, since it is likely that most forest soils are in the range of Al buffering (cf. section 1.1). Finally the content of important heavy metals, i.e. Pb, Cd, Zn, Cu, Cr and Ni were determined in level 2 plots for the humus layer in 1990 and even for the complete soil profile in 1992 and 1995. This is done since heavy metal pollution, which is known to occur in forest soils (Kleijn et al., 1989), might also affect forest vitality.

In the soil solution, all the major cat ions and anions were measured, i.e. H, Al, Fe, Ca, Mg, K, Na,  $NH_4$ ,  $NO_3$ ,  $SO_4$ , Cl and  $RCOO^-$ .

#### Analysing methods

Total contents of C and N were determined by wet oxidation according to the methods of Kurmies (Kurmies, 1949) and Kjehldahl (Hesse, 1971), respectively. Total S contents were extracted in a concentrated mixture of nitric acid and hydrochloric acid and analyzed by atomic absorption spectrometry (AAS). Total contents of P and of Ca, Mg, K, Zn, Cu, Cr and Ni in the humus layer were extracted in a concentrated mixture of sulphuric acid and nitric acid and analyzed by inductively coupled plasma atomic emission spectrometry (ICP). Total contents of Pb and Cd were determined by an extraction with concentrated (9%) hydrochloric acid during three hours followed by ICP analyses of the extract.

Exchangeable contents of Al, Fe, Ca, Mg, K and Na were measured by extraction with a 0.01 M solution of silver thiourea during four hours (Chabra et al., 1975) followed by analyses with ICP (Al, Fe, Ca and Mg) and atomic absorption spectrometry (AAS). Exchangeable  $NH_4$  contents were measured by extraction with 1.0 M KCl (Coleman et al., 1959) followed by analyses with a colorimetric technique (flow injection analyzer; FIA). The CEC was determined from the decrease in Ag concentration before and after the extraction (Ag is measured by ICP) and the exchangeable H content was calculated from the difference in CEC and exchangeable cat ion content.

The readily dissolvable contents of Al and Fe were measured by extracting the samples during four hours in the dark with an acid ammonium oxalate solution at pH 3 followed by ICP analyses of the extract (Schwertmann, 1964). In this extract the P content was also measured.

Dissolved concentrations of major ions were determined by centrifugation of a soil sample of 400 g. at 7500 rpm during 20 minutes. The centrifuge tube was made of polyoxymethylene (deldrin) and consisted of an upper soil-holding cup with a perforated base (holding ca 250 ml of soil) and a lower solution holding cup, fitting in a 500 ml hole rotor. Centrifugation generally took place within one day (18-30 hours) after collection of the soil samples. Immediately after centrifugation the pH was measured.

The soil solution samples were filtered over 0.45  $\mu$ m. Concentrations of Al, Fe, Ca, Mg and SO<sub>4</sub> were analyzed by ICP, K and Na by AAS and NH<sub>4</sub>, NO<sub>3</sub>, H<sub>2</sub>PO<sub>4</sub> and Cl by FIA. The pH was measured by means of potentiometry. The concentration of organic anions was calculated from the DOC content (Oliver et al., 1983) that was measured by an organic carbon analyzer. More information on the approaches is given in de Vries and Leeters (2001) and in Leeters et al. (1998).

Year	Num	Layer	Pa	ram	eters	s Ar	nalys	sed															
	ber of plots		5	Si:	$\mathbf{AI}$	Fe	Са	Ζ	Х	Ζ	Ζ	Ν	Ζ	Ū	S	Р	0	C	0	Ν	Р	U	Ż
1990	12	0 - 30	х	х	х	х	х	х	х	х	х	х	Х	х	х	х	х						
	12	60 - 100	х	х	х	х	х	х	х	х	х	х	х	х	х	х	х						
1992	12	0-10	х	х	x	x	х	х	х	х	х	х	x	х	х	х							
	12	10 - 30	х	х	х	х	х	х	х	х	х	х	х	х	х	х							
	12	30 - 60	х	х	х	х	х	х	х	х	х	х	х	х	х	х							
	12	60 - 100	x	х	х	х	х	х	х	х	х	х	х	х	х	х							
1993	12	0 – 10	х	х	x	x	x	x	х	х	х	x	x	х	х	x		x	x	x	х	x	
1770	12	10 - 30	x	x	x	x	x	x	x	x	x	x	x	x	x	x		x	x	x	x	x	
	12	30 - 60																					
			X	X	X	x	x	x	X	x	x	x	X	x	x	x		X	X	X	X	X	
	12	60 - 100	х	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х	
1994	12	humus	х	х	х	х	х	х	х	х	х	х	х	х	х	х							
	12	0 - 10	х	х	х	х	х	х	х	х	х	Х	х	х	х	Х							
	12	10 - 30	х	х	х	х	х	х	х	х	х	х	х	х	х	Х							
	12	30 - 60	х	х	х	х	х	х	х	х	х	х	х	х	х	х							
	12	60 - 100	х	Х	х	х	х	х	х	х	х	х	х	х	х	х							
1995	14	humus	х	х	х	х	х	х	х	х	х	х	x	х	х	х		х	x	х	х	x	х
	14	0 - 10	х	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х	х
	14	10 - 30	х	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х	х
1996	12	humus	х	х	x	x	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х	х
	12	0 - 10	x	x	x	x	x	x	x	x	x	x	x	x	x	x		x	x	x	x	x	x
	12	10 - 30	x	x	x	x	x	x	x	x	x	x	x	x	x	x		x	x	x	x	x	x
	12	30 - 60	X	x	X	x	x	x	x	x	x	x	x	x	x	x		x	x	X	x	x	x
	12	60 - 100	x	X	x	X	x	x	X	X	X	x	X	X	X	X		X	X	X	X	X	X
1997	14	humus	х	х	x	x	x	x	х	х	x	x	x	х	x	x			x	х			
1777	14	0 - 10																					
	14		X	X	X	X	X	x	X	X	X	x	X	X	X	X			X	X			
		10 - 30	X	X	X	х	x	x	X	X	X	x	X	X	X	X			X	X			
	14	30 - 60	х	х	х	х	х	х	х	х	х	х	х	х	х	х			х	х			
	14	60 - 100	х	х	х	х	х	х	х	х	х	х	х	х	х	х			х	х			
1998	14	humus	х	х	х	х	х	х	х	х	х	х	х	х	х	х			х	х			
	14	0 - 10	х	х	х	х	х	х	х	х	х	х	х	х	х	х			х	х			
	14	10 - 30	х	х	х	Х	х	х	х	х	х	Х	х	х	х	Х			х	х			
	14	30 - 60	х	х	х	х	х	х	х	х	х	х	х	х	х	Х			х	х			
	14	60 - 100	х	х	х	х	х	х	х	х	х	х	х	х	х	х			х	х			
1999	14	humus	х	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х		х	х	х
	14	0 - 10	х	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х		х	х	х
	14	10 - 30	х	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х		х	х	х
	14	30 - 60	х	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х		х	х	х
	14	60 - 100	х	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х		х	х	х
2000	14	humus	х		x	x	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х	х
	14	0 - 10	x		x	x	x	x	x	x	x	x	x	x	x	x		x	x	x	x	x	x
	14	10 - 30	X		x	x	x	x	x	x	x	x	X	x	x	x		x	X	X	x	X	x
	14	30 - 60	х		х	х	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х	х
	14	60 - 100	х				х		х		х												
	1-1	00 - 100	Λ		Х	х	л	Х	л	Х	л	Х	Х	Х	Х	Х		Х	Х	Х	Х	х	Х

Table 2.8 Layers and analysed soil solution parameters over the period 1990 - 2001

Year	Num	Layer	Param	eter	s Ar	nalys	sed															
	ber of plots		c Si	$\mathbf{M}$	Fe	Са	Μ	Х	Z	Z	Μ	Z	CI	S	Р	0	$\mathbf{C}_{\mathbf{r}}$	Cu	Ζ	Pb	Cd	ž
2001	7	humus	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х	х
	7	0 - 10	х	х	х	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х	х
	7	10 - 30	х	х	х	х	х	х	х	х	х	х	х	х	x		х	х	х	х	х	х
	7	30 - 60	х	х	х	х	х	х	х	х	х	х	х	х	x		х	х	х	х	х	х
	7	60 - 100	X	х	х	х	х	х	х	х	х	х	х	х	х		х	х	х	х	х	х

# Second period 2003 - 2006

#### Installation and materials

In 2002 during the months May and June, tension lysimeters were installed in the plots 106 Hardenberg, 1040 Zeist and 2085 Dwingeloo. On these plots deposition measurements also take place. The first lysimeters data were available in the last quarter of 2002, but these samples were not fit for use in the data series. The first year in which the soil solution could be collected all year round is 2003. In June 2005 the lysimeters 12 and 14 of plot 2085 Hardenberg were replaced on both depths. The tension lysimeters are made of polyacrylaat. Since they are, unlike ceramic material which are in use by most countries, inert for the influence of heavy metals. Polyacrylaat also does not influence the measurements of Ca, DOC and CEC. The polyacrylaat lysiometers are made and tested according to the protocol recorded in the *Standaard werkvoorschrift E3200 van Alterra*.

A total of 60 tension lysimeters are placed in each plot, 30 in 15 m and 30 on 80 cm below surface. They are placed according to the protocol as described in the *Standaardwerkvoorschrift E3202* from Alterra. Sampling takes place according to the protocol described in the *Standaardwerkvoorschrift E3203* also from Alterra. Soil solution is extracted using glass bottles which are brought to a diastolic pressure of 0.8 bar. These bottles are connected to the tensio lysimeters for one day and disconnected the next day (mostly 24 ours between).

# Spatial sampling design

In order to get an accurate estimate of average concentrations in the soil solution for the plot area and over the measured time period a special sampling strategy was designed. More than just one (mixed) sample was analyzed in order to get a better estimated of average concentrations of the soil solution. This also makes it possible to give information about the uncertainty of the measurement.

Sampling locations have been selected by stratified simple random sampling. The strata are geographical strata (blocks) of equal area formed by the *k*-means clustering algorithm (Brus et al., 1999). In this method the plot is divided into a large number of square cells (pixels). These cells are then clustered using the x- en y-coordinates of the centres of the cells as classification variables. The centroids of the resulting clusters are finally used to calculate the Thiessen polygons. In each stratum six locations are randomly selected. The order of selection is registered. The soil solution collected at the locations of the same order is used to form a composite aliquot, i.e. the water is mixed. With six locations per stratum this results in six composites. Note

that due to the stratification the soil solution of a given composite is collected at locations that are well spread over the entire plot. This makes that the composite mean is an unbiased and precise estimate of the plot mean. If we would have taken only one composite, we would not be able to estimate the sampling variance of the estimated. As explained hereafter, by taking more than one composite, we have replicates from which we can estimate the sampling variance of the mean. Figure 2.5 gives a schematic representation of the plot, the division in strata and the locations were a sample is taken.

We first consider the case in which there is no non-response. In this case an unbiased estimator for the plot average is the unweighted average of the concentrations measured on the composites (De Gruijter et al., 2006):

$$\hat{\overline{y}} = \frac{1}{n} \sum_{i}^{n} y_{i}, \qquad (2.1)$$

where  $y_i$  is the concentration of composite *i*, and *n* is the number of composites. As we have six independent realizations of the sampling design, one can use these six samples to estimate the sampling variance:

$$\hat{v}(\hat{\bar{y}}) = \frac{v(y_i)}{n} = \frac{1}{n(n-1)} \sum_{i}^{n} \left( y_i - \hat{\bar{y}} \right)^2$$
(2.2)

In case of no non-response, estimator (2.1) is unbiased because the strata have equal area, and as a result the sample is self-weighting.

For some plots no soil solution could be collected at one or more sampling locations. In sampling theory this is referred to as non-response. Due to this non-response the number of aliquots (soil solution sample at an individual location) of the composites varies. In Figure 2.5 only composite 1 consist of five aliquots, one aliquot from each stratum, whereas composite 2, 4 and 6 consist of four aliquots and composite 3 and 5 of three aliquots. The problem is that only the concentration of composite1 is an unbiased estimate of the mean concentration of the plot. For composites 2, 4, 6 one stratum is not represented in the composite, for composite 3 and 5 two strata are absent. As a result the concentrations of the individual composites are not unbiased estimates of the plot mean. From this it follows that, in the presence of nonresponse, the unweighted average of the composite concentrations is not an unbiased estimate of the plot mean. Therefore an unbiased estimator of the plot-mean concentration and an estimator for its standard deviation was developed that can be used in situations with non-response. In case of non-response a linear regression approach is followed to estimate the plot average. The idea behind this estimation procedure is that the concentrations of the composite samples can be used to estimate the mean concentration for each stratum (or for groups of strata). The mean concentration for the plot is then estimated by the (un)weighted average of the estimated stratum means. This procedure is explained in more detail in Appendix 4.

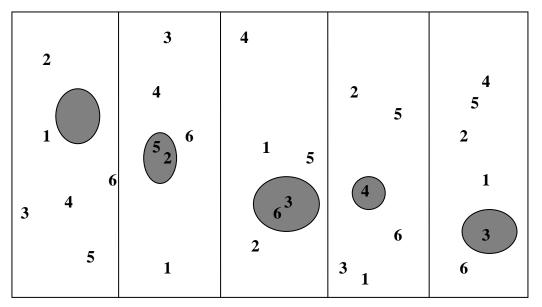


Figure 2.5 Schematic representation of composite sampling across strata in the presence of nonresponse. Aliquots taken at locations of equal number are grouped to form one composite. Shaded areas are areas where no soil solution could be collected (non-response).

#### Sampling periods

In the ICP Forests Manual, Part III Submanual on Soil Solution Collection and Analysis, it is suggested to combine the sampling of deposition and soil solution and collect the samples monthly. In this case, however, sampling times are not chosen at fixed moments but related to the precipitation surplus. Precipitation data from nearby weather stations over the period 1970 - 1999 are used to run the SWAP model and calculate daily precipitation surplus over a 30 years period. SWAP (Soil, Water, Atmosphere and Plant) simulates transport of water, solutes and heat in unsaturated/saturated soils (Kroes & van Dam, 2003) and (Van Dam, 2000).

The plots 106 Hardenberg and 2085 Dwingeloo both are located within 20 km of the weatherstation Heino and plot 1040 Zeist is located within 10 km of the weather station De Bilt. Both stations, Heino and De Bilt, are monitored by the Royal Netherlands Meteorological Institute (KNMI). The values of these stations are considered representative for the plots. Table 2.9 shows the average yearly precipitation surplus for the above mentioned plots.

Table 2.9 Average yearly precipitation surplus over the period 1970 - 1999 and precipitation surplus per sampling period for the level 2 plots where soil solution is measured

Plot	Name	Average yearly precipitation surplus (mm) over the period 1970 - 1999	Precipitation surplus (mm) per sampling period (12 per year)
106	Hardenberg	252.5	21.0
1040	Zeist	356.7	29.7
2085	Dwingeloo	282.3	23.5

Dividing the average yearly precipitation surplus into 12 even parts, every part has a precipitation surplus per sampling period as given in Table 2.9. Within the first part a starting value for the precipitation randomly selected (e.g. for plot 1040 Zeist a value between 1 and 297, 195 was picked.). New starting values are picked every new year. These values determines when the first sampling takes place (in our example on the day that the average yearly precipitation surplus has reached 19.5 mm). Next sampling takes place when the same amount of precipitation surplus is reached (in our example on the day that the precipitation surplus is 19.5+29.7=49.2 mm) The advantage of this approach compared to stratified sampling is that sampling moments can not be to close to each other. For practical sampling only takes place on Mondays, Tuesdays, Wednesdays and Thursdays on the first possible day that the cumulative precipitation surplus is not yet exceeded. For practical reasons also, a cluster is made of the plots 106 Hardenberg and 2085 Dwingeloo. Calculated precipitation surplus for these plots corresponded well. Therefore the day averages of this plots were averaged and these values were used to determine the sampling days. This makes it possible to sample both plots on the same day.

Due to weather conditions or practical reasons some sampling periods were not successfully. In January and February 2003 no samples could be taken due to freezing conditions and in October 2003 due to drought conditions. The 24<sup>th</sup> of December sampling was not conducted because of Christmas holidays. The samplings planned on 29 July, 19 November and 11 December were not executed for plot 1040 Zeist because of misunderstanding. In 2004 all 12 planned samplings were executed but in December, plot 2085 Hardenberg, did not deliver soil solution samples due to frost.

#### Laboratory measurements

In the field 60 bottles with soil solution are gathered. The are marked with codes corresponding to the sampling point and depth. Collected samples are brought directly to the laboratory by car, where they are stored in a cold storage room (4 °C). The same day in the laboratory the amount of ml is written down for each bottle. Samples are mixed over the strata. All first points of the five strata are joined into on sample, all second points of the five strata are also joined into one sample and so on. The samples are mixed with equal amounts of every sample point. Therefore the point with the least soil solution determines the total amount of the mixed sample. Mixed samples contain 100 ml at most. In total when soil solution was obtained at all points, 6 mixed samples per depth are analysed, usually this is less.

Analyses of pH, N-NO<sub>3</sub>, N-NH<sub>4</sub> and DOC is preformed direct after mixing. Other parameters (see Table 2.10) are analyses normally within a period of 6 weeks. Analyses of the samples from January 2003 until mid November 2004 were carried out by the Alterra laboratory. Analyses of samples taken after mid November 2004 were carried out by the Chemical and Biological Soil Laboratory of Wageningen University, using the same methods as before.

Analyses carried out were sometimes restricted by the total amount of soil solution (in ml) available. 40 ml is sufficient to carry out all the desired analyses. But in case the total amount is less than 40 ml choices were using a priority sequence as shown in Table 2.10. Results of analysed parameters were checked for extreme values of the parameters separately. Checked is also whether the Na/Cl ratio is between 0.7 and 1.3, whether P ortho < P total and whether the difference between the sum of catand anions is less than 20%. These checks were carried out for the mixed samples. Values below the detection limit are included in the calculation using a value equal to half the detection limit. If the calculated average value was below the detection limit the values was reported as -1.

Parameter	Unit	Status	< 10	10-20	20-30	30-35	35-40	> 40
			ml	ml	ml	ml	ml	ml
pН		mandatory	Х	Х	Х	Х	Х	Х
DOC	[mg/l]	mandatory						
DON	[mg/l]	optional						Х
TC/NPOC	[mg/l]			Х	Х	Х	Х	Х
K	[mg/l]	mandatory			Х	Х	Х	Х
Mg	[mg/l]	mandatory			Х	Х	Х	Х
Ca	[mg/l]	mandatory			Х	Х	Х	Х
Al	[mg/l]	mandatory			Х	Х	Х	Х
S	[mg/l]	mandatory			Х	Х	Х	Х
Na	[mg/l]	optional +			Х	Х	Х	Х
Zn	[µg/l]	optional +			Х	Х	Х	Х
Cu	[µg/l]	optional +			Х	Х	Х	Х
Fe	[mg/l]	optional			Х	Х	Х	Х
Mn	[mg/l]	optional			Х	Х	Х	Х
Ptot	[mg/l]	optional			Х	Х	Х	Х
Cr	[µg/l]	optional			Х	Х	Х	Х
Ni	[µg/l]	optional			Х	Х	Х	Х
Pb	[µg/l]	optional			Х	Х	Х	Х
Cd	[µg/l]	optional			Х	Х	Х	Х
NO <sub>3</sub> -N	[mg/l]	mandatory		Х	Х	Х	Х	Х
NH4-N	[mg/l]	optional +		Х	Х	Х	Х	Х
Cl	[mg/l]	optional +				Х	Х	Х
P-PO <sub>4</sub>	[mg/l]	-					Х	Х

Table 2.10 Priority sequence for analyses related to the amount of sample available

#### 2.2.5 Crown condition

The condition of the Dutch forest was observed nation wide on 1500 (being a subset of 3000) sites each year over the period 1984 – 1994, specifically in view of the potential adverse effect of air pollution. The aim of this national inventory was to gain insight in the development of the vitality of major tree species in the Dutch forests. The vitality was judged in view of the crown condition, more specifically in terms of defoliation and discolouration. This information however, can only be used to register changes in vitality and is not sufficient to show cause-effect relations between the threats and the forest condition. In 1995 the new Forest Health Monitoring Network started under the responsibility of the Ministry of Agriculture, Nature Management and Fisheries and was executed by The National Reference Centre for Nature Management (IKC-N, presently known as DK). This Forest Health Monitoring Network covered 200 forest stands and was initiated to get a better insight in the effects of acidification, eutrophication and desiccation on the forest ecosystem. The 14 level 2 plots are part of this Forest Health Monitoring Network. This Forest Health Monitoring Network however has stopped in 2000. Crown condition now is monitored only on the level 1 and level 2 plots in the context of the ICP (see section 2.1).

Crown condition is observed on 25 marked trees in the plot and expressed by means of defoliation and discolouration. Defoliation is observed in percentages loss of leaves for Oak and in percentages occupation for needles in regard to a supposedly normal situation. Defoliation for conifers is later on determined by the assumption that a normal healthy Douglas-fir has a full needle occupation of 5 years and Scots pine, older than 40 years, has a full needle occupation of 2 years. For the reporting of defoliation the actual percentage is used round off in steps of 5%, for instance 0 = 0%; 5 = >0 - 5%; 10 = >5 - 10% etc. Discolouration is observed as (i) a percentage of the surface of the needles or leaves and (ii) a percentage of the surface of the crown totally. These percentages are reported in 6 classes: 1 = 0%, 2 = 1 - 10%, 3 = 11 - 25%, 4 = 26 - 60%, 5 = >60% and 6 = dead, as described in Hilgen & Reuver (1996). Later on the discolouration of needles and leaves and the discolouration of the crown is joined (see Table 2.11) into one discolouration expressed and reported to the EU in 4 classes:

- 0. no discolouration (0 10%)
- 1. slight discolouration (>10 25%)
- 2. moderate discolouration (>25 60%)
- 3. severe discolouration (>60%)

Table 2.11 Discolouration class as combination of discolouration of needles and leaves and discolouration of the crown used for reporting to the EU

Discolo	uration crown	Discolourati	on need	les and leave	S			
Class	Percentage	Class	1	2	3	4	5	6
		Percentage	0	1 - 10	11 - 25	26 - 60	>60	dead
1	0		0	0	0	0	0	
2	1 - 10		0	0	0	0	0	
3	11 - 25		0	0	1	1	0	
4	26 - 60		0	1	2	2	2	
5	>60		0	1	2	3	3	
6	dead							9

Table 2.11 is applied for submission of the data to the EU since 2000. Before 2000, we have no information about the discolouration of needles and leaves and the crown separately.

Besides defoliation and discolouration observations were made for possible damage and if reducible what causes the damage. For instance game and grazing, insects, fungi, direct action of man, fire, known local or regional pollutant or other damage. Identified causes of damage were reported with a 1 if the damage was recognized and more than 10%; with a 0 if the damage was not recognized and with a blank if no assessments were made. Assessments of damage by insects or fungi were made as the percentage of damage to the total needle- or leaf mass using the classes: 1 = 0%, 2 = 1 - 10%, 3 = 11 - 25%, 4 = 26 - 60%, 5 = >60%. Damage was reported starting class 3 or higher, so if it was more than 10%.

From 2002 on the assessment for the level 2 plots was extended with the mandatory parameters removals and mortality (14 classes), social status (4 classes), crown shading (6 classes) and visibility (4 classes) and also with the optional parameters flowering and fruiting (both 3 classes).

From 2004 onward more information is required about the damage causes in order to provide more information about their impact on crown condition. The assessment of the damage causes has extended to a symptom description, a determination of the cause and a quantification of the symptoms. Therefore the parameters specification of the effected part, symptom specification, location in crown, cause, cause name and extent must be assessed. However, these parameters were not assessed in 2004 and neither in 2005 because we discovered this desired change in reporting when submitting the data of 2004 in December 2005. In order to make submission possible we estimated the new parameters as truthful as possible using the notes that were made during the assessment.

In the Netherlands the condition of the forest is not reported by defoliation and discolouration but by vitality, also expressed in 4 classes. The vitality class is a combination of the defoliation and discolouration (Table 2.12). In this case defoliation is expressed in 4 classes: 1 = 0 - 10%, 2 = 11 - 25%, 3 = 26 - 60% and 4 = >60%. The combination of discolouration of needles and leaves and the discolouration of the crown in the Netherlands is not done according to Table 2.11 but according to Table 2.12.

Discolou	uration crown	Discolouration needles and leaves						
Class	Percentage	Class	1	2	3	4	5	
	_	Percentage	0	1 - 10	11 - 25	26 - 60	>60	
1	0		0	0	0	0	0	
2	1 - 10		0	1	1	1	2	
3	11 – 25		0	1	2	2	2	
4	26 - 60		0	2	3	3	4	
5	>60		0	2	4	4	4	

Table 2.12 Discolouration class as combination of discolouration of needles and leaves and discolouration of the crown used to establish the vitality class in the Netherlands

Defoliation class	Discolouration class								
	1	2	3	4					
1	1	1	2	3					
2	2	2	3	3					
3	3	3	4	4					
4	4	4	4	4					

Table 2.13V itality class as combination of defoliation and discolouration

The vitality class is derived for all 25 trees in a plot where defoliation and discolouration is observed. The vitality of the forest stand is established as the average vitality class of those 25 trees. The vitality classes can be described as:

- 1. Vital forest: This forest shows all characteristics, as expected based on the tree species and age and considered normal for the local situation. There is no doubt about the expectations for the future.
- 2. Less vital forest: This forest shows characteristics on which the expectation for the future is good. There are however some symptoms that indicate that the future expectations can also be negative.
- 3. Hardly vital forest: This forest shows characteristics on grounds of which the future expectations are bad. This forest however still has the possibilities to become vital again.
- 4. Non vital forest: This forest has on grounds of the characteristic to be considered lost or is dead already.

#### 2.2.6 Growth

Growth parameters as diameter at breast height and tree height were first measured in 1995 and repeated in 2000 and 2005. The diameter at breast height (dbh) was measured by using tape and tree height by using Blume-Leiss Suunto hypsometers. The first measurement was performed by IKC-LNV at the start of the level 2 monitoring. At the same time, the coordinates of the plots and the coordinates of the separate trees were determined (See also Section 2.1). For the second measurement, all trees were numbered so that identification was possible and the data of both measurements could be submitted. The numbers 101 - 549 were given to the trees in the upper (A) part of the plot and the numbers 550 - 999 were given to the trees in the lower (B) part of the plot (The numbers 1 - 100 were reserved for the crown condition survey). The plot was split into two parts to ease the field measurements. Table 2.14 gives an overview of the number of measured tree per plot and their distribution over tree classes. The class division comes from a national classification, the so called HOSP. It shows that most trees are classified as co-dominant or suppressed. In this class also most of the height measurements are preformed. In 2000 the classification of the separate trees was not updated there fore the numbers per class remained similar. In 2005 the classification was updated so a different distribution over the classes is possible. Note that it is also possible that the number of measured trees in 2005 is higher than in 2000 caused by the fact that in 2000 some trees were not found and therefore not measured. For plot 61, 61 of the 101 trees were not found. Therefore this plot is revisited in 2001 by another fieldworker who completed the measurements.

Plot	Year	Numł	per of t	rees n	neasu	red f	for dia	meter	at		Numl	ber o	of tree	es m	easu	ired	for	heig	ht
		breast	heigh	t (1.3m	n) in e	each	class				in eac	h cla	iss*						
		total	1	2	3	4	5	6	7	8	total	1	2	3	4	5	6	7	8
39	1995	157	1	114	20	4		18			10		10						
	2000	93		72	17	3		1			9		9						
58	1995	255		123	10	1	121				22		22						
	2000	162		84	3		75				18		17			1			
61	1995	106		76	2		26	2			12		12						
	2000	101		73	2		24	2			15		15						
82	1995	229	5	181	33			2	8		20	1	19						
	2000	205	5	164	28			1	7		0								
106	1995	60		54	3		2	1			8		8						
	2000	54		51	3						9		9						
	2005	58	6	40	6	2	4				34	6	18	4	2	4			
129	1995	121	4	88	3		23	3			18	2	16						
	2000	101	4	77	3		17				18	2	16						
174	1995	140		139	1						22		22						
	2000	115		114	1						16		16						
175	1995	126	107	19							19		19						
	2000	86	68	18							17		17						
	2005	65		63	2						26		26						
226	1995	298	2	231	63			2			20		20						
	2000	198	2	170	25			1			13	2	11						
1012	1995	136		108	25			3			18		18						
	2000	131		107	24						18		18						
1040	1995	95		81	5		8	1			10		10						
	2000	93		81	4		8				8		8						
	2005	92		75	10		7				24		24						
2080	1995	128	1	102	25						17		17						
	2000	86	1	66	19						14		14						
2084	1995	304		252	49			2			43		43						
	2000	169		144	25						30		30						
	2005	166	4	137	23			1			41	2	38				1		
2085	1995	114		101			5	8			15		15						
	2000	96		96							13		13						
	2005	101		91	3		7				28		27	1					

Table 2.14 The number of measured trees per plot and their distribution over tree classes

\*) 1=dominant, 2=co-dominant, 3=controlled, 4=suppressed, 5=ingrowth, 6=dead, 7=over- or upper tree, 8=hanging

As Table 2.14 shows, height was only measured for part of the trees, mostly representative trees of the co-dominant class. For plot 82 it was not possible to measure height in 2000, because of the density of the forest. In stead of a measurement the average height of this plot was estimated to be 16 - 18 m. For the other plots the height of the trees that were not measured was calculated using the percentage growth in height from the measured trees in regard to the previous sampling period according to:

(((height (t) - height (t-1))/height (t-1))\*100) (2.3)

The average of this percentage growth per plot or per tree class (when measurements were available in more classes) was used as factor (n%) to calculate the height of the non-measured trees according to:

Height (t) = height (t-1) + (height (t-1)\*n%/100)

#### 2.2.7 Ground vegetation

The ground vegetation was sampled in 2000 and in 2005. In 2000, all 14 plots were included, but in 2005 only five were sampled (0106, 0175, 1040, 2084 and 2085). Relevés were made according to the method of Braun-Blanquet (1961), using an adapted cover scale (Table 2.15). This scale is identical to the scale used in the 'Meetnet Functievervulling' (Dirkse et al., 2007), and in the 'Meetnet Vitaliteit en Verdroging' (Van Dobben et al., 1997; Van Dobben & de Vries, 2001). In each plot, four circular subplots with a diameter of 4.89 m were laid out, giving a total area of 300 m<sup>2</sup>. The subplot layout is given in Figure 2.6. The centre point of each plot was marked in the field by means of a buried magnetic marker. The vegetation in each subplot was subdivided in layers according to Table 2.16, and cover estimates per species were made in each layer. A consequence of the layer definition in Table 2.16 is that a single species can appear in more than one layer! Vascular species, mosses, liverworts and lichens were included; cryptogams were always sampled and their identifications were checked afterwards. Note that both plot size and layer definitions are slightly different from what is now prescribed in the Manual!.

Ground cover			
Cover Code	Cover scale	Cover (%)	
1	< 0,1 %	0.1	
2	0,1 - 1 %	0.5	
3	1 - 5 %	3	
4	5 - 10 %	8	
5	10 - 25 %	18	
6	25 - 50 %	38	
7	50 - 75 %	68	
8	75 - 90 %	83	
9	90 - 100%	95	

Table 2.15 Cover scale used in the assessment. Cover is 'true' cover i.e. the percentage of the soil covered by living plant parts in perpendicular projection

Table	e 2.16	Layer	definition

Vegetation	Definition
layer	
Tree layer	Everything higher than 6 m
Shrub layer	Woody vegetation between 1.5 and 6 m
Herb layer	Woody vegetation below 1.5 m, and all non-woody vascular
	plants
Moss layer	All cryptogamic plants

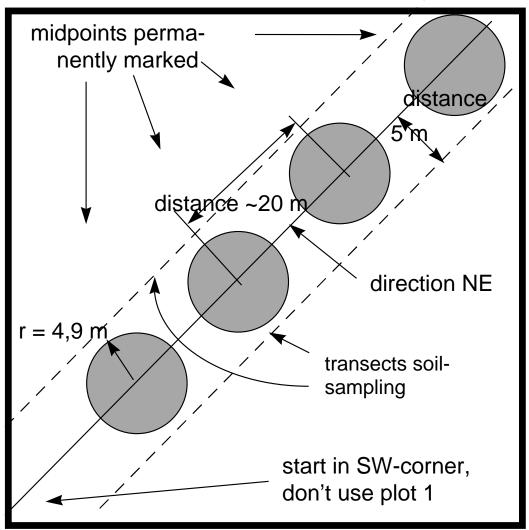


Figure 2.6 The layout of a subplot for ground vegetation assessment.

## 2.3 Data storage

Over the years data were stored in separate (text or excel) files and managed by different people. Part of this study was to collect all data and store them together in one national database. This database was build in Microsoft Access and contains of a front end and a backend. The backend contains the data originally sampled in the field or analysed in the laboratory. This backend is only accessible for one or two data managers so it is unlikely that users can damage the data. The front end is the user interface which is made very easy to use. Knowledge of access is not required. The user only has to press buttons or chose from pop-up menu's to get an overview of the data or an explanation of the used definitions. It is also possible to get the data in the formats that are required for data submission to the EU. These formats have changed over the years. The formats are always corresponding to the actual situation in the concerning year.

# 3 Atmospheric deposition

As mentioned in chapter 2, atmospheric deposition was measured at four locations, Dwingeloo, Hardenberg, Speuld and Zeist. Measurements started in 1995 but were incomplete for 1996 and 1997, complete measurements around the year were conducted from 1997 onwards.

#### 3.1 Calculation of total atmospheric deposition

Total deposition was calculated on the basis of data for bulk deposition and throughfall, according to a slightly adapted canopy budget model developed by Ulrich (1983) and extended by Bredemeier (1988) and by Draaijers and Erisman (1995). Data were available for the year 1995 and then for the years 1997-2004 onwards. The quality of the data in the years 1996 and 1997 was insufficient for a reliable calculation.

In the canopy budget model, annual total deposition is derived by correcting the input by both throughfall and stem flow for exchange processes, occurring within the forest canopy. Since stem flow data were missing at all plots, the annual stemflow was estimated from the annual throughfall according to Ivens (1990):

$$X_{sf} = X_{tf} \cdot \alpha / (1 - \alpha) \tag{3.1}$$

where X is a given ion (H, Ca, Mg, K, Na, NH<sub>4</sub>, NO<sub>3</sub>, SO<sub>4</sub>, Cl), sf is stemflow  $(mol_c.ha^{-1}.yr^{-1})$ , tf is throughfall  $(mol_c.ha^{-1}.yr^{-1})$  and  $\alpha$  is an empirical value

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

$$\begin{array}{ll} \alpha = 0.24 & \text{age} < 20 \\ \alpha = 0.31 - 0.0034 \cdot \text{age} & 20 < \text{age} < 90 \\ \alpha = 0.0 & \text{age} > 90 \end{array} \tag{3.2}$$

For deciduous forests,  $\alpha$  was set at 0.12 independent of age. The same values were used for coniferous forests with an unknown age (Ivens, 1990). More information is given in De Vries et al. (1999).

Total deposition fluxes of base cat ions were calculated according to (Ulrich, 1983):

$$BC_{td} = \frac{Na_{tf} + Na_{sf}}{Na_{bd}} \cdot BC_{bd}$$
(3.3)

where BC is Ca, Mg, K, td is total deposition and bd is bulk deposition (both in  $mol_c.ha^{-1}.yr^{-1}$ )

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Eq. (3.3) is based on the assumption that (i) Na does not interact with the forest canopy (inert tracer) and (ii) the ratios of total deposition over bulk deposition are similar for Ca, Mg, K and Na. Specifically in coastal areas, this assumption is not always valid. Canopy leaching induced by the internal cycle of these nutrients, was thus computed by the difference between the sum of BC in throughfall and stemflow minus total deposition according to:

$$BC_{ce} = BC_{tf} + BC_{sf} - BC_{td}$$
(3.4)

where ce is canopy exchange ( $mol_c$ .ha<sup>-1</sup>.yr<sup>-1</sup>). Canopy exchange of  $SO_4^{2-}$  is assumed negligible. The total deposition of this ion was thus calculated as:

$$SO_{4,td} = SO_{4,tf} + SO_{4,sf}$$

$$(3.5)$$

 $NH_4$  and H interact with the forest canopy by exchange with base cations (Roelofs et al., 1985). We assumed that the total canopy uptake of H<sup>+</sup> and  $NH_4^+$  is equal to the total canopy leaching of Ca<sup>2+</sup>, Mg<sup>2+</sup> and K<sup>+</sup> taking place through ion exchange, corrected for the leaching of weak acids. The  $NH_4$  throughfall and stemflow flux was thus corrected for canopy uptake to calculate the total deposition of  $NH_4$  according to (After Van der Maas et al., 1991; Draaijers & Erisman, 1995):

$$NH_{4,td} = NH_{4,tf} + NH_{4,sf} + NH_{4,ce}$$

$$(3.6)$$

with:

$$NH_{4,ce} = BC_{ce} - WA_{ce} - H_{ce}$$
(3.7)

and:

$$H_{ce} = \left(\frac{H_{tf} \cdot xH}{NH_{4,tf} + H_{tf} \cdot xH}\right) \cdot \left(BC_{ce} - WA_{ce}\right)$$
(3.8)

where xH is an efficiency factor of H in comparison to  $NH_4$  and WA is weak acids. Based on experiments in the laboratory (Van der Maas et al., 1991), it was assumed that H<sup>+</sup> has per mol an exchange capacity six times larger than  $NH_4^+$  (xH = 6). The estimation of the weak acid concentration was based on the sum of  $HCO_3^-$ , derived from the pH and an assumed atmospheric CO<sub>2</sub> pressure, and RCOO<sup>-</sup> derived from DOC or the difference in concentration of cat ions minus strong acid anions (see Annex 4). The weak acid canopy exchange was calculated as:

$$WA_{ce} = WA_{tf} + WA_{sf} - 2WA_{bd}$$
(3.9)

The total deposition of protons was calculated as:

$$H_{td} = H_{tf} + H_{sf} + H_{ce}$$

$$(3.10)$$

Total deposition of  $NO_3^-$  was calculated according to:

$$NO_{3,td} = NO_{3,tf} + NO_{3,sf} + NO_{3,ce}$$
(3.11)

where the canopy exchange of  $NO_3^-$  equals the canopy exchange of nitrogen minus the canopy exchange of  $NH_4^+$ . The canopy exchange of nitrogen was calculated according to:

$$N_{ce} = NH_{4,ce} \cdot \left(\frac{NH_{4,tf} \cdot xNH_4 + NO_{3,tf}}{NH_{4,tf} \cdot xNH_4}\right)$$
(3.12)

where  $NH_{4,ce}$  is calculated according to Eq. (3.12) and the  $xNH_4$  is an efficiency factor of  $NH_4^+$  in comparison to  $NO_3^-$  (we assumed that  $xNH_4 = 6$ ). Actually, Draaijers and Erisman (1995), assumed canopy uptake of  $NO_3^-$  to be negligible. There is, however, ample evidence that this is not true, specifically since  $NO_3$  in throughfall is often less than  $NO_3$  in bulk deposition in low deposition areas.

#### 3.2 Atmospheric deposition between 1995 – 2005

Figure 3.1 shows the temporal variation in deposition for four sites as derived from throughfall measurements and bulk deposition data after application of the canopy ex-change model. Deposition of  $SO_4$  appears to decrease substantially over the period 1995-2005 most clearly noticeable from 2000 onwards (Figure 3.1A). The sites at Hardenberg and Zeist show a more or less steady decline since 1995, whereas the sites at Dwingeloo and Speuld show more variation with a peak deposition in 1998. Furthermore, all sites show a small increase in 2003, followed by a decrease in deposition since that time.

Deposition levels of total N, potential acid and base cations show more erratic variation over the years 1995-2005 with possibly a marginal decrease for total N and potential acid (Figure 3.1B, C). Adverse effects of acidifying compounds' can be mitigated by deposition of base cat ions (Ca, Mg and K). The deposition of base cations in Hardenberg is nearly twice that at other locations and shows large temporal variation between 1995 and 2005. Base cation depositions at other locations are substantially lower and differ only marginally with lowest levels in Zeist followed by Speuld and Dwingeloo. These locations also show less temporal variation than Hardenberg (Figure 3.1D).

Variations over the years are mainly due to varying meteorological circumstances. Deposition of  $SO_4$ , total N and potential acid are highest for Speuld and lowest for Zeist, whereas deposition levels at Dwingeloo and Hardenberg differ only marginally and have intermediate levels over the period 1995 -2005 (Figure 3.1).

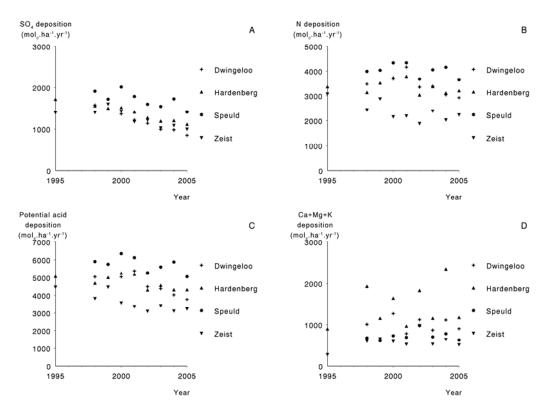


Figure 3.1 Deposition of SO<sub>4</sub>, total N, potential acid and base cat ions at the locations Dwingeloo, Hardenberg, Speuld and Zeist over the period 1995-2005

As with total N deposition, deposition of both  $NO_3$  and  $NH_4$  are highest for Speuld and lowest for Zeist while Dwingeloo and Hardenberg are in between and differ only slightly (Figure 3.2). There are no clear trends in the deposition of  $NO_3$  and  $NH_4$ . The N/S ratio clearly increases with time, in line with the decrease in S deposition at all plots, while the  $NH_4/NO_3$  ratio tends to stay constant with the exception of an increase for Zeist.

#### 3.3 Trends in atmospheric deposition

To determine if trends in atmospheric deposition at monitoring locations are significant, simple linear regression was conducted. An overall trend, assuming that the change at all locations is equal with different deposition levels for individual locations, was determined according to:

$$Y_{deposition} = a_0 + a_1. \text{ year } + a_2 \text{ location}$$
(3.13)

where Y deposition is the deposition of element Y in  $mol_cha^{-1}yr^{-1}$  and year is calendar year AC. Regression coefficients, standard errors and the number of significant coefficients (a2) for different locations are presented in Table 3.1. The value of  $a_1$ gives the trends in time and is significant when it is larger than twice the standard deviation of  $a_1$ .

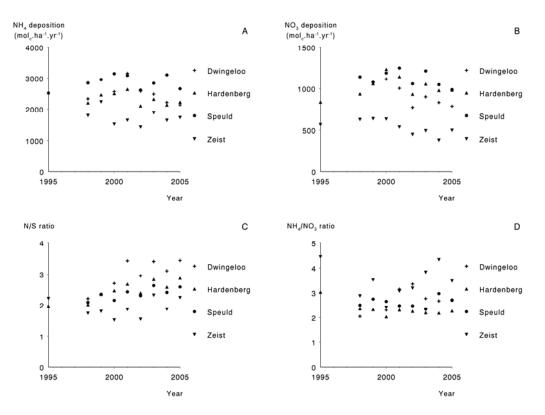


Figure 3.2 Deposition of NH<sub>4</sub>, NO<sub>3</sub> and the ratio's NH<sub>4</sub>/NO<sub>3</sub> and N/S at the locations Dwingeloo, Hardenberg, Speuld and Zeist over the period 1995-2005

Comparison of the value of  $a_1$  and its standard deviation shows that the deposition of SO<sub>4</sub> has a significant decrease of 64 mol<sub>c</sub>ha<sup>-1</sup>yr<sup>-1</sup>. Deposition of total N shows a slightly less clear but still significant decrease of 61 mol<sub>c</sub>ha<sup>-1</sup>yr<sup>-1</sup>. Deposition of potential acid, being the summed deposition of total-N and SO<sub>4</sub>, thus shows a significant decrease of 125 mol<sub>c</sub>ha<sup>-1</sup>yr<sup>-1</sup>. The deposition of total-N split up into NH<sub>4</sub> and NO<sub>3</sub> shows a significant decrease for both of respectively 47 and 14 mol<sub>c</sub>ha<sup>-1</sup>yr<sup>-1</sup>. Deposition of base cations shows a slight increase but this increase is not significant. Absolute deposition levels for total-N, SO<sub>4</sub> and potential acid vary significant with the location but apparently show a more or less similar and significant decrease.

Due to the slightly larger decrease of S compared to N, the N/S ratio shows a small but significant increase. The larger decrease of  $NH_4$  compared to  $NO_3$ , causes the  $NH_4/NO_3$  ratio to decrease slightly but this change is not significant.

Element	$a_0$	a <sub>1</sub>	se a <sub>1</sub>	se obs.	R <sup>2</sup>	nr of plots with a <sub>2</sub> significant
SO <sub>4</sub>	129994	-64	7.0	108	86	3
Total N	125278	-61	19	294	82	3
Potential acid	255271	-125	22	336	86	3
BC	-44041	23	19	294	57	3
$\rm NH_4$	95638	-47	16	248	73	3
$NO_3$	29640	-14	7.1	109	82	3
N/S ratio	-133.1	0.068	0.017	0.26	73	4
NH <sub>4</sub> /NO <sub>3</sub> ratio	12.87	-0.005	0.029	0.45	46	2

Table 3.1 Trends in overall annual deposition (mol<sub>o</sub>.ha<sup>1</sup>.yr<sup>-1</sup>) and in deposition ratios (-) in the period 1995-2005

The overall decrease of  $SO_4$  deposition appears to differ per location as shown in Table 3.2 where trends are estimated for individual locations according to:

 $Y_{deposition} = a_0 + a_1$ . year

Table 3.2 Trends in annual SO<sub>4</sub>, NO<sub>3</sub> and NH<sub>4</sub> deposition (mol<sub>2</sub> $ha^{1}$ .yr<sup>1</sup>) in the period 1995-2005

Element	Location	<b>a</b> 0	a1	se a <sub>1</sub>	$R^2_{adj}$
SO <sub>4</sub>	Dwingeloo	205788	-102	5.7	98
	Hardenberg	126276	-62	5.1	95
	Speuld	122849	-61	22	50
	Zeist	104752	-52	15	58
Total N	Dwingeloo	219371	-108	59	28
	Hardenberg	46569	-22	32	*
	Speuld	82138	-39	40	*
	Zeist	184575	-91	31	48
NH4	Dwingeloo	106627	-52	58	*
	Hardenberg	64747	-31	20	15
	Speuld	49112	-23	32	*
	Zeist	145552	-72	31	35
$NO_3$	Dwingeloo	112743	-56	14	72
	Hardenberg	-18178	10	14	*
	Speuld	33027	-16	14	4
	Zeist	39023	-19	7.8	39

All locations show a significant decrease of  $SO_4$  deposition but the magnitude of the decrease is about twice as high for Dwingeloo (102 mol<sub>c</sub>ha<sup>-1</sup>yr<sup>-1</sup>) as for other locations. Estimates of trends in nitrogen compounds for individual locations result in low fits and a non significant trend as shown in Table 3.2, except for Zeist where a significant decrease is observed in total N, NH<sub>4</sub> and NO<sub>3</sub> and in Dwingeloo where a significant decrease is observed for total N and NO<sub>3</sub>. For other locations trends for the different Nitrogen compounds are not significant despite the significant overall decrease seen in Table 3.1. This is probably due to the limited number of measurements and the erratic temporal variation per location preventing the identification of significant trends at individual locations, while overall trends based on more measurements shows significant decreases.

# 4 Crown condition and chemical composition of foliage and soil solution

# 4.1 Crown condition

#### 4.1.1 Crown condition between 1984 – 2006

Figure 4.1 shows the trends in crown condition per tree species at plot level, in terms of a change in vitality class, over the period 1984–2006. As described in Section 2.2.5, 1 stand for Vital forest, 2 for Less vital forest, 3 for Hardly vital forest and 4 for Non vital forest. Occasional estimates on vitality before 1990 seem higher than later estimates and are therefore disregarded in the further analysis because of an expected discrepancy in assessment methodology. From 1990 onwards, the following trends can be discerned:

- For Scots pine there is a great fluctuation in vitality over the years. There is clear recovery at all level I plots, but at the level 2 plots, there has been a decline around 1993 and there are no signs of recovery until 2006.
- For Douglas fir the forest condition gets worse around 1993 and there are no signs of recovery until 2006.
- The condition for Oak seems to become worse over the years. There is a slight change from class 2 (less vital forest) around 1990 towards class 3 (hardly vital forest) in 2006.

For all tree species there is apparently no direct relation between the deposition level and the vitality. Vitality is the reflection of many influences and it is not clear on this moment what causes the low vitality and its erratic temporal variation. In section 4.1.2 regression analysis is performed to determine if significant trends exist in the defoliation and discoloration, which determine the vitality class.

## 4.1.2 Trends in crown condition

The estimation of temporal trends using linear regression is not suitable when the dependant variable is defined in classes, such as tree vitality. Therefore discoloration and defoliation, which determine the vitality class as described in section 2.2.5, were separately used in the trend analysis. Both defoliation and discoloration are recorded in percentage classes that are not uniform in class width; therefore the class number is not a suitable dependant variable in regression analysis. The original percentage classes were therefore transformed to the middle of class and this percentage for defoliation and discoloration was used as dependant variable in the trend analysis. Regression models for all locations and tree species were fitted according to:

$$Defoliation = a_0 + a_1 \text{ year} + a_2 \text{ tree species}$$
(4.1)

Discoloration =  $a_0 + a_1$  year +  $a_2$  tree species (4.2)

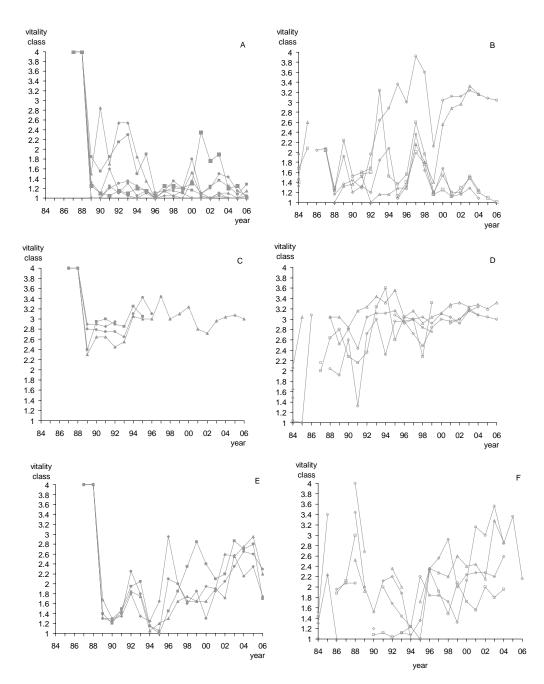


Figure 4.1 Vitality class per plot over the period 1984 – 2006 for Scots pine (A and B), Douglas-fir (C and D) and oak (E and F) subdivided at level 1(A, C and E) and level 2(B, D and F) plots

Trend analysis for defoliation (Table 4.1) shows a small but significant increase of 0.45% per year. The crown discoloration also increases marginally but significantly with 0.18% per year. Surprisingly the discoloration of the leaves shows an opposite trend and decreases significantly with 0.23% per year. Absolute levels of defoliation and discoloration differ significantly between all three tree species apparent from the 3 significant absolute levels in Table 4.1.

Element	<b>a</b> <sub>0</sub>	a <sub>1</sub>	se a <sub>1</sub>	R <sup>2</sup>	nr of tree species with a <sub>2</sub> significant
Defoliation	-888	0.451	0.034	32	3
Discoloration of the leaves	500	-0.232	0.060	17	3
Discoloration of the crown	-352	0.181	0.039	7	3

Table 4.1 Coefficients for overall temporal trend models with different absolute levels per tree species

Separate regression models per tree species were also fitted with the possibility of different absolute levels for each location according to:

Defoliation =  $a_0 + a_1$  year +  $a_2$  location

Discoloration =  $a_0 + a_1$  year +  $a_2$  location

Trends in defoliation, per tree species (Table 4.2), reveals that defoliation of Oak increases very significant with 0.77% per year and for 5 out of 9 plots the absolute defoliation levels also differ significantly. There is also a large overall increase in defoliation of Douglas fir (1.24%/yr) that is just significant and again the absolute defoliation levels differ significantly for 5 out of 9 plots. Pine shows no significant change in defoliation.

Table 4.2 Trend models for defoliation and discoloration per tree species

Characteristic	Tree species	$a_0$	a <sub>1</sub>	se a <sub>1</sub>	R <sup>2</sup>	nr of plots with a2 significant
Defoliation	Pine	-174	0.089	0.046	28	10/11
	Douglas	-2434	1.242	0.062	20	5/9
	Oak	-1519	0.770	0.060	9	5/9
Discoloration	Pine	406	-0.183	0.143	2	
of the leaves	Douglas	716	-0.356	0.087	1	
	Oak	-229	0.121	0.107	1	
Discoloration	Pine	-761	0.382	0.074	3	
of the crown	Douglas	-473	0.238	0.044	5	
	Oak	-616	0.315	0.109	3	

The overall decrease in discoloration of needles and leaves seen in Table 4.1 is caused by the significant decrease for Douglas (-0.36%/yr) fir; Oak and Pine show no significant change in discoloration of needles and leaves (Table 4.2). Absolute levels of discoloration of needles and leaves do not differ significantly between plots. The overall increase in discoloration of the crown seen in Table 4.1 is caused by a significant increase for all three tree species (Table 4.2). Absolute levels of discoloration of the crown do not differ significantly between plots.

The increase in defoliation and discoloration of the crown is contrary to an expected decrease based on trends in atmospheric deposition. Only Douglas-fir shows a significant decrease in discoloration of the needles that may be related to decreasing atmospheric deposition. Other factors such as plagues, diseases or droughts are probably more important when explaining observed trends and differences between tree species and locations.

# 4.2 Chemical composition of the foliage

#### 4.2.1 Chemical composition of the foliage between 1990 – 2005

#### Data analysis in view of thresholds for chemical composition of the foliage

The chemical composition of foliage was first measured in 1990. In the period 1992 - 1997 it was measured yearly and since 1999 every second year. In total, this gives us ten values of the analysed elements over a period of 13 years for four or five sites per tree species. Some elements however are not analysed consistently over the years. For an overview of the elements analysed over the years: see section 2.2.2. From the elements reported in this chapter sulphate was not analysed in 1990 and 1996. Each data point in the graphs of *Figure 4.2* is an average of data from four or five plots of the same tree species. The National data set for 1990 and 1995 consists of an average of around 16 - 27 plots for Douglas-fir, 42 plots for Scots pine and 28 - 51 plots for Oak.

In this section we compare the results for foliar content with the European standards for Scots pine and Oak (Stefan et al., 1997). These standards are somewhat lower than the Dutch standards in the guidelines for forest fertilization (Van den Burg & Schaap, 1995). A European standard for Douglas-fir is lacking. We used the European standard for Spruce for Douglas-fir as well. We describe trends, remarkable developments and deviations from the normal pattern.

The nutrient condition of Oak, Scots pine and Douglas-fir is described in relation to the European standards for low and high levels of the nutrient concentration (Stefan et al., 1997), also mentioned in De Vries et al. (2000). These thresholds are developed in a European Expert Committee in regular meetings. In Table 4.3 an overview is given of the differences for some major elements: P, K, Mg and N. The low threshold levels in the EU standard are lower than the Dutch threshold levels. The high threshold levels are often in the same order of magnitude, resulting in usually larger optimal intervals in the EU standard. Douglas-fir thresholds are not available in the EU standard. Douglas-fir is rather comparable to Spruce in the Dutch standard. For potassium and magnesium, the Dutch threshold values are exactly the same for Spruce and Douglas-fir. For the nitrogen and the phosphorus content, the values are only slightly different. Therefore, we have used the EU standards for Spruce to describe the nutrient condition of Douglas-fir in the Netherlands. High levels of nitrogen and low levels of the other elements are most important for the interpretation of the forest condition in the Netherlands and are indicated in bold in Table 4.3

Values between the low and the high thresholds in Table 4.3, lead to optimal growth. Nutrient deficiencies occur below the low level. For nitrogen, levels much higher than the high threshold are excessive, and lead to anomalies like a high arginine content, an amino acid with a high nitrogen content, in e.g. Douglas-fir and Corsican pine (Van Dijk et al., 1992; Van Dijk, 1993). This is associated with higher risks for diseases'.

Table 4.3 Comparison of Dutch and EU standards for threshold values (mg.g<sup>1</sup>) for low and high nutrient content (based on V an den Burg & Schaap, 1995; Stefan et al., 1997). In the Dutch thresholds, spruce is Norway and Sitka spruce, pine is Scots pine. In bold, high levels of nitrogen and low levels of the other elements in the EU standards.

Tree species		Ν		р		K		Mg	
1		EU	NL	EU	NL	EU	NL	EU	NL
Spruce	Low	<12	<13	<1.0	<1.4	<3.5	<6.0	<0.6	< 0.7
	High	>17	>17	>2.0	>2.0	>9.0	>8.0	>1.5	>1.0
Douglas-	Low	-	<14	-	<1.4	-	<6.0	-	<0.7
fir	High	-	>18	-	>2.2	-	>8.0	-	>1.0
Pine	Low	<12	<14	<1.0	<1.4	<3.5	<5.0	<0.6	< 0.7
	High	>17	>18	>2.0	>1.7	>10	>7.0	>1.5	>1.0
Oak	Low	<15	<23	<1.0	<1.4	<5	<6.0	<1.0	<1.6
	High	>25	>28	>1.8	>1.7	>10	>8.0	>2.5	>2.8

The Dutch low threshold levels for element content are between 40 and 60% higher than the EU standard for phosphorus and potassium, for all three species, and in magnesium, for Oak (see Table 4.3). The differences in the low thresholds in other cases are small (less than 15% higher). Also the high threshold for nitrogen is comparable in the Dutch and the EU standards for all three species. The reasons for these differences are unknown.

Recently, a field test has been done to evaluate the effects of forest fertilisation, with phosphorus, potassium and magnesium, and liming, as subsidised by the Dutch government. Fertilisation is subsidized when levels for P, K and Mg are too low (pure element content, or ratios like in Figure 4.3) and liming is possible when the soil pH-KCl is below 3.2 (Olsthoorn & Wolf, 2006). The main results were that the effect of fertilisation and liming could not be found in nutrient contents and soil pH, because of the large variation in the data. Another result of the evaluation was that growth was rather satisfactory, also before the fertilization and liming. Most tree species have grown between 2 and 3 mm in the annual rings, leading to a diameter increment of 4 to 6 mm per year. The diameter increment was used as an indicator of vitality, as the vitality parameters foliage retention and colour were not measured before the fertilization or liming. This growth level seems to indicate that there is no real vitality problem at the Dutch threshold levels for P, K and Mg. Even a pH-KCl below 3.2 does not seem to be a great problem. The conclusion in the Evaluation report of Olsthoorn & Wolf (2006) is that the threshold levels for a low content of P, K and Mg could be stricter. The EU standard is stricter, so might be a better indication of real vitality problems for the trees. This underlines our choice to use the EU standards in this 10 years monitoring report.

#### Results

Figure 4.2 shows the average nutrient level for the measurements in Dutch forest stands from 1990 onwards. For N and S, only the thresholds for a high content are indicated in the graphs. For the other elements only the low threshold is relevant for

the nutrient condition. The National data in 1990 and 1995 are taken from a larger data set (from 150 to 200 stands in total with a variety of tree species, see section 2.2.2). Figure 4.2A shows that almost all values from the Level 2 plots are above the high nitrogen threshold. This indicates an increased risk for stress induced by drought, frost and diseases. However, the N concentrations show a declining trend and near 2005, they reach the threshold for all tree species. The data for Douglas-fir and Oak from the large national dataset are well in range with the four selected plots for the level 2 monitoring.

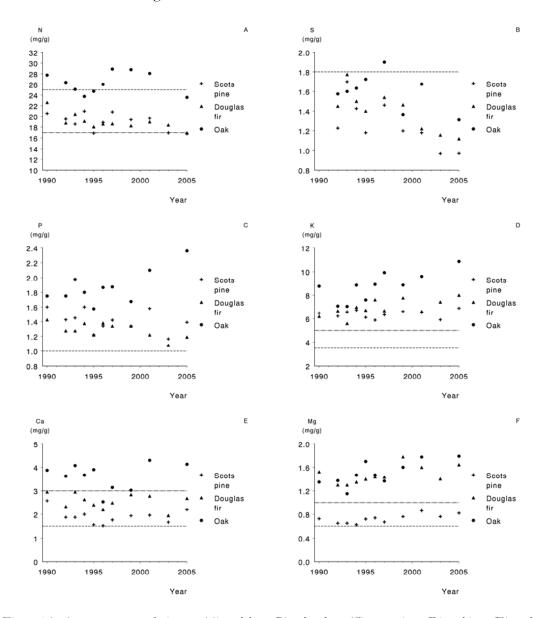


Figure 4.2 Average contents of nitrogen (A), sulphate (B), phosphate (C), potassium (D), calcium (E) and Magnesium (F) in foliage for the tree species Douglas Fir, Oak and Scots pine over the years 1990 – 2005. Lines in the figure are EU thresholds for high levels of nitrogen and low levels of the other elements as given in bold in Table 4.3.

Sulphur (Figure 4.2B) is always under the threshold for high levels (a threshold for Oak is lacking in the EU standard). The sulphur content shows large annual variation, as can be seen in the nineties, but the plot indicates a strong declining trend. Phosphorus (Figure 4.2C) never scores below the low threshold for all three tree species. In 1990 and 1995, the Level 2 plots are in line with the National dataset.

The potassium content is always above the threshold for low levels. The annual variation is rather large for Oak. The calcium content is also above the threshold for low levels, apart from Oak, where 1996 and 1999 have a low level, around the threshold. For all tree species, the levels remain fairly constant, rather close to the low threshold. The calcium and magnesium content for Scots pine is near the low threshold. Oak and Douglas-fir are well above the low threshold for both Ca and Mg. The fluctuations are fairly large, with no clear visible trend.

If we would compare these graphs with the Dutch threshold levels for low and high nutrient content (see Table 4.3), this description would be completely different for some elements. The high threshold for nitrogen would be somewhat higher than the EU threshold for Scots pine and Douglas-fir. Therefore, the levels would be less over the threshold. There is no Dutch standard in the National guidelines for sulphur content. The low threshold for phosphorus is the same for Oak, Scots pine and Douglas-fir, namely 1.4 mg.g<sup>-1</sup>. This would mean that nearly all Douglas-fir data would become too low, and most of the Scots pine data. Oak still would be well above the low phosphorus threshold in the Dutch standard. The potassium content for Oak and pine would still be adequately above the low threshold, but Douglas-fir would be near the low threshold, and score too low in 1993. For calcium there would be no changes for Oak and Scots pine (same levels as in the EU standard, not shown in Table 4.3). For Douglas-fir, it means that nearly all years would score below the low threshold for calcium (2.5 mg.g<sup>-1</sup>). For magnesium, the difference would be small for Douglas-fir, still well above the low threshold. The difference would be large for Oak and Scot pine, as Scots pine would fall below the threshold for most of the years. Only the last few years would be above the low threshold. As the annual variation in Oak is large for magnesium, around half the years would score below the low threshold. The last years would be just above the low threshold.

Figure 4.3 shows the trends in the ratio of nitrogen with phosphorus, potassium, magnesium and calcium, respectively. As the availability of nitrogen in the Netherlands is quite high, the element concentrations compared are quite often near the standards for an unbalanced nutrition, sometimes with signs of deficiency symptoms.

For oak all ratios are always below the high threshold level, except for N/Ca in the period 1996 to 1999. This means there is only a problem with the Ca levels in some years. Maybe the pH of the soil is too low for satisfactory calcium availability. This is in line with the element contents in Figure 4.2, which are satisfactory, except for a high nitrogen content and sometimes low calcium levels.

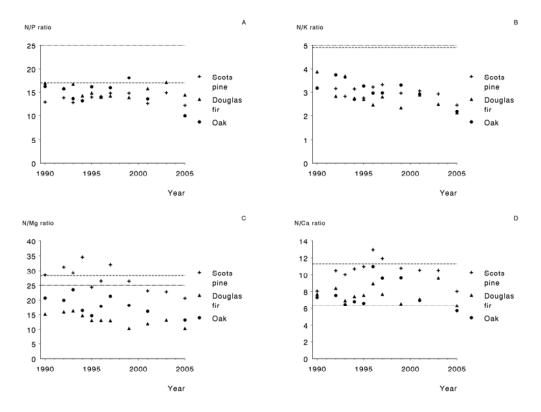


Figure 4.3 Average nutrient ratio's of N and P (A), N and K (B), N and Mg (C) and N and Ca (D) in foliage for the tree species Douglas Fir, Scots pine and Oak over the yeas 1990 - 2005.

Douglas-fir is mostly just under, but quite close to the high threshold or slightly over it: for N/P in 1990, 1993 and 2003, never for N/K, for never for N/Mg, and never for N/Ca. However, the ratio of nitrogen with the other elements is very high, and should preferably decrease. There is no sign of a decease so far. Scots pine is more at risk than Douglas-fir through its high N/Mg and N/Ca ratios. In the nineties, there are many years that the value is above the high threshold. Scots pine is not so close to the high threshold for N/P and N/K.

Table 4.4 evaluates combinations of balanced and unbalanced with sufficient or insufficient levels of nutrient contents of each nutrient (from De Vries, 2000). For stands in the right bottom corner, both ratios and contents are satisfactory. This is the best situation, indicated with 1. For stands in the top left corner, both the ratios and the contents are unsatisfactory. This is the worst situation, indicated with 4.

Table 4.4 Allocation of results to different classes with respect to nutrient availability and nutrient balance. The numbers indicate the order of the four classes.

	Insufficient nutrient availability	Sufficient nutrient availability
Unbalanced nutrient status	4 ratio high	3 ratio high
Balanced nutrient status	<ul><li>concentration low</li><li>ratio low/average</li><li>concentration low</li></ul>	concentration average/high 1 ratio low/average concentration average/high

Table 4.5 lists the percentages of Level II plots in the four different classes with respect to nutrient availability and nutrient balance (see Table 4.4) as a function of tree species. Results are averages for all years, so a trend cannot be seen in this table.

Table 4.5 Percentages of plots in four different classes, see Table 4.4, with respect to nutrient availability and nutrient balance as a function of tree species. The percentage per element and tree species is a result of the combination of four to five plots and ten years.

Tree		Р		Κ		Ca		Mg		All	
		Insu	Suff								
Df	Unbal.	2	19	0	2	0	4	0	0	2	21
	Bal.	0	79	0	98	0	96	0	100	0	77
Sp	Unbal.	2	4	0	0	13	29	13	25	25	44
1	Bal.	0	94	0	100	2	56	0	62	2	29
Oak	Unbal.	0	3	0	0	28	46	6	3	31	43
	Bal.	0	97	0	100	0	26	0	91	0	26
All	Unbal.	2	9	0	1	12	25	6	10	19	35
	Bal.	0	89	0	99	1	62	0	84	1	45

Results show that (on average over the whole 10 year period) phosphorus and potassium are always sufficiently available. Nearly all stands fall in the class of sufficient and balanced fertilization. Only Douglas-fir has 19% stands that are not balanced with nitrogen, as nitrogen is high. The calcium nutrition is a problem for Scots pine and Oak, with low numbers in the highest class (below 60%) and a substantial number in the lowest class. Magnesium nutrition mainly is a problem for Scots pine (13% in class 4, and only 62% in class 1) and not all for Douglas-fir.

The main problem for Douglas-fir is the sometimes unbalanced phosphorus nutrition due to high nitrogen contents (19% of the plots). For Oak the main problem is the often insufficient and/or unbalanced calcium nutrition (46% of the plots), whereas the main problem for Scots pine is the sometimes unbalanced calcium and magnesium nutrition (25-30% of the plots).

## 4.2.2 Trends in chemical composition of the foliage

To explore if there is a significant trend in nutrient contents in foliage over time, several linear-regression analyses were conducted with year, plot and tree species as explanatory variables. A preliminary check was conducted whether the nutrient contents show auto covariance, meaning that the measured contents in one year depends on the values of previous years. This was not the case thus satisfying an important requirement for the use of linear regression. The nutrient content in the foliage was used as dependant variables while, at first, year and tree species were used as explanatory variables, according to:

 $Y-content = a_0 + a_1 \text{ year} + a_2 \text{ tree species}$ (4.3)

where Y-contents represents the content of element Y in mg  $g^{-1}$  in the foliage,  $a_1$  represents the change in Y-content per year and  $a_2$  represents the different absolute levels for each tree species. Secondly separate regression models were fitted for each tree species according to:

$$Y-content = a_0 + a_1 year + a_2 location$$
(4.4)

where  $a_2$  represents the different absolute levels for each location.

Results of the first analyses (Table 4.6) show that N-content and S-content in the foliage decrease significantly over the period 1990-2005. The decrease of N-content and S-content in the foliage is significant for Douglas-fir and Pine but not for Oak (Table 4.7). The N contents in Oak (27 mg.g<sup>-1</sup>) are significantly higher than those in Douglas-fir or Scots pine (both 19 mg.g<sup>-1</sup>) see also Figure 4.2. The difference in S contents between tree species is also significant. Oak (1.6 mg.g<sup>-1</sup>) has the highest S contents followed by Douglas-fir (1.4 mg.g<sup>-1</sup>) and Scots pine (1.3 mg.g<sup>-1</sup>).

Table 4.6Linear regression results for element concentrations with dependant variables year and tree speciesThe value of  $a_1$  gives the trends in overall element concentrations in foliage (mg.kg<sup>1</sup>) in the period 1990-2005

Element	$a_0$	a <sub>1</sub>	se a <sub>1</sub>	se obs	R <sup>2</sup>	nr of species with a <sub>2</sub> significant
Ν	246.	-0.1135	0.0529	2.41	66	2
S	63.5	-0.03117	0.00588	0.231	39	3
Р	12.7	-0.00567	0.00575	0.262	41	3
Κ	-150.2	0.0784	0.0216	0.984	48	3
Ca	35	-0.0166	0.0140	0.638	54	3
Mg	-38.59	0.01969	0.00440	0.201	77	3

K-content and Mg-content in the foliage increase significantly over the period 1990-2005 and also show a significant difference in absolute levels between tree species. Overall changes in P-content and Ca-content are not significant but the differences in absolute levels between tree species are (Table 4.6). The K content in Douglas-fir is significantly higher (6.9 mg.g<sup>-1</sup>) than in Scots pine (6.4 mg.g<sup>-1</sup>) and the highest of all is Oak (8.6 mg.g<sup>-1</sup>). Mg content is highest in Douglas-fir (1.5 mg.g<sup>-1</sup>) and Oak (1.5 mg.g<sup>-1</sup>) which both are significantly higher than those in Scots pine (0.7 mg.g<sup>-1</sup>).

Results (Table 4.7) show that the increase of Mg-content in the foliage is significant for all tree species, while the increase in K-content is only significant for Douglas (Table 4.7). The absolute level of Mg-content in the foliage differs significantly between 2 plots for Douglas and between 4 plots for Pine and Oak. The differences between the tree species are significant; Ca content is highest for Oak (3.6 mg·g<sup>-1</sup>), less for Douglas-fir (2.6 mg·g<sup>-1</sup>) and least for Scots pine (1.9 mg·g<sup>-1</sup>). P contents for Scots pine (1.4 mg·g<sup>-1</sup>) is significantly higher than for Douglas-fir (1.3 mg·g<sup>-1</sup>) and in Oak (1.8 mg g<sup>-1</sup>) is significantly higher than in both Scots pine and Douglas-fir. For none of the three tree species a significant change is detectable over the period 1990-2005 (Table 4.7). Oak is maintaining a rather high level for phosphorus. The absolute level of P-content in the foliage for Pine and Douglas differs significantly between 3 plots and for Oak between 4 plots.

Element	Tree species	<b>a</b> 0	<b>a</b> <sub>1</sub>	se a <sub>1</sub>	R <sup>2</sup>	nr of plots with a <sub>2</sub> significant
Ν	Pine	329.	-0.1557	0.0734	9	1
	Douglas	429.	-0.2052	0.0918	24	2
	Oak	-62	0.043	0.109	15	3
S	Pine	74.7	-0.03678	0.00798	34	1
	Douglas	75.6	-0.03714	0.00725	40	1
	Oak	24.1	-0.0114	0.0201	-	1
Р	Pine	16.2	-0.00737	0.00593	37	3
	Douglas	13.8	-0.00629	0.00593	56	3
	Oak	-33.6	0.0175	0.0127	47	4
K	Pine	-17.3	0.0116	0.0191	33	3
	Douglas	-185.8	0.0966	0.0306	20	1
	Oak	-223.	0.1153	0.0601	37	4
Ca	Pine	30.7	-0.0146	0.0129	39	3
	Douglas	13.8	-0.0056	0.0176	19	1
	Oak	4.5	-0.0006	0.0413	23	2
Mg	Pine	-16.85	0.00883	0.00288	65	4
5	Douglas	-39	0.02038	0.00736	26	2
	Oak	-62	0.03163	0.00991	51	4

Table 4.7 Linear regression results for element concentrations per tree species with dependant variables year. The value of  $a_1$  gives the trends in element concentrations in foliage (mg.kg<sup>1</sup>) per tree species in the period 1990-2005

#### 4.3 Chemical composition of the soil solution

#### 4.3.1 Composition of soil solution between 1990 – 2001

As described in paragraph 2.2.4 the chemical composition of the soil solution is collected from centrifuged soil samples from 1990-2001 and collected with lysimeters from 2003-2006. Because of methodological difference and the length of first measurement period only the measurements from 1990-2001 were used for trend analysis. Measurements from 2003 – 2006 were used to calculate nutrient budgets as described in chapter 6. In acidic soils, such as those at all Level 2 plots, atmospheric deposition of S and N compounds leads to elevated Al concentrations, in response to elevated concentrations of sulphate (SO<sub>4</sub>) and nitrate (NO<sub>3</sub>), and also to accumulation of NH<sub>4</sub> in situations where nitrification is (strongly) inhibited. This may cause nutrient imbalances, since the uptake of base cation nutrients (Ca, Mg, K) is reduced by increased levels of dissolved Al and NH<sub>4</sub> (Boxman et al., 1988).

Average concentrations of major ions in the soil solution for 1990-2001 of the topsoil (0-30 cm) and subsoil (30-60 cm) per tree species are given in Table 4.8. Changes in concentrations of  $SO_4$ ,  $NH_4$   $NO_3$ , which respond most directly to changes in S and N deposition, and in Al, Ca and Mg, which are major indicators for soil acidification, for the period 1990-2001 are shown in Figure 4.4 for the topsoil and in Figure 4.5 for the subsoil.

Table 4.8 Average concentrations in the soil solution of the topsoil and subsoil between 1990 and 2001 per tree species.

	Tree	рΗ	Al	Fe	Ca	Mg	Κ	Na	$\mathrm{NH}_4$	$NO_3$	Cl	$SO_4$	$\mathrm{PO}_4$
Тор	Sp	4.3	0.41	0.04	0.36	0.15	0.12	0.48	0.16	0.37	0.58	0.60	0.004
soil	Df	3.7	0.86	0.03	0.52	0.30	0.14	0.95	0.25	0.71	1.18	1.02	0.005
	Oak	4.0	0.33	0.03	0.34	0.18	0.23	0.33	0.12	0.62	0.45	0.40	0.014
	All	4.0	0.54	0.03	0.41	0.21	0.16	0.60	0.17	0.56	0.74	0.68	0.007
Sub	Sp	4.4	0.39	0.01	0.31	0.14	0.07	0.47	0.06	0.36	0.51	0.58	0.001
soil	Df	4.2	0.90	0.05	0.49	0.25	0.08	0.95	0.09	0.64	1.16	0.97	0.001
	Oak	4.4	0.29	0.03	0.30	0.16	0.10	0.35	0.08	0.38	0.40	0.44	0.003
	All	4.3	0.53	0.03	0.37	0.18	0.08	0.59	0.08	0.46	0.69	0.67	0.002

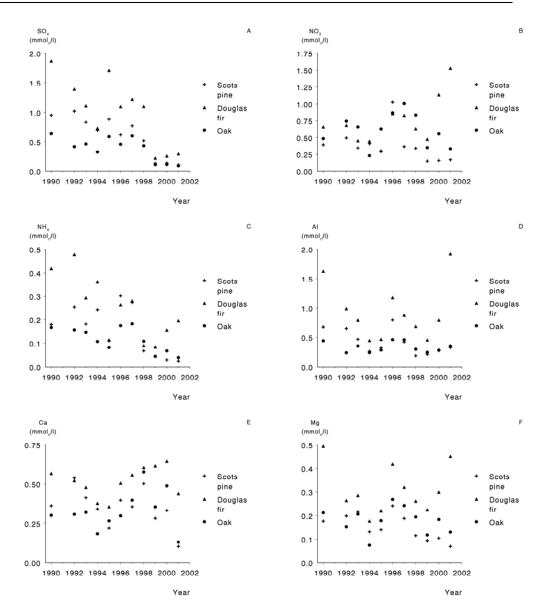


Figure 4.4 Average soil solution concentrations of sulphate (A), nitrate (B), ammonium (C), aluminium (D), calcium (E) and Magnesium (F) in the topsoil (0-30 cm) for the tree species Douglas Fir, Oak and Scots pine over the years 1990 - 2001.

 $SO_4$  concentrations in the soil solution of the topsoil (Figure 4.4A) are comparable to those in the subsoil (Figure 4.5A).  $SO_4$  concentrations are lowest under Oak and highest under Douglas-fir. Since 1999,  $SO_4$  concentrations are substantially lower than in previous years.  $NO_3$  concentrations in the soil solution of the topsoil (Figure 4.4B) are higher than of the subsoil (Figure 4.5B). In 2000 and 2001  $NO_3$ concentrations in the topsoil are about twice as high as in previous years.  $NO_3$ concentrations in the topsoil are generally lowest under Scots pine and alternating highest under Douglas-fir and Oak.  $NO_3$  concentrations in the subsoil are generally lowest under Oak and highest under Douglas-fir.

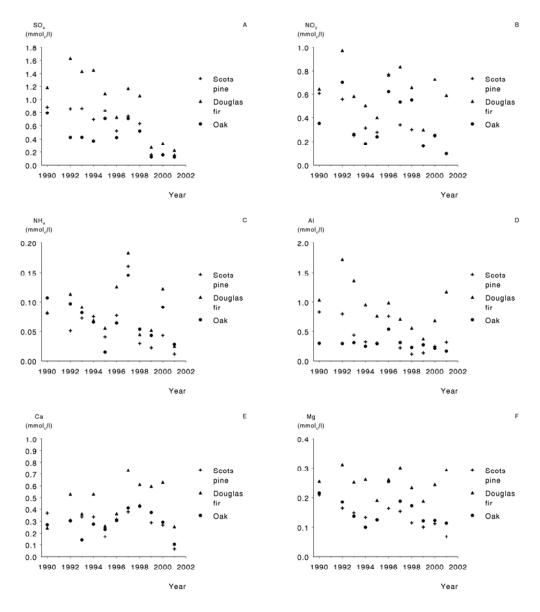


Figure 4.5 Average soil solution concentrations of sulphate (A), nitrate (B), ammonium (C), aluminium (D), calcium (E) and Magnesium (F) in the subsoil (60-100 cm) for the tree species Douglas Fir, Oak and Scots pine over the years 1990 - 2001.

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 $NH_4$  concentrations in the soil solution of the topsoil (Figure 4.4C) are higher than of the subsoil (Figure 4.5C). In 1997  $NH_4$  concentrations in the soil are about twice as high as in previous and preceding years.  $NH_4$  concentrations in the topsoil are generally lowest under Oak and highest under Douglas-fir.  $NH_4$  concentrations in the subsoil are generally lowest under Scots pine and highest under Douglas-fir.

Al concentrations in the soil solution of the topsoil (Figure 4.4D) are about the same as in the subsoil (Figure 4.5D). Al concentrations in both the topsoil and subsoil are generally lowest under Oak and highest under Douglas-fir. Ca concentrations in the soil solution of the topsoil (Figure 4.4E) are about the same as in the subsoil (Figure 4.5E). Ca concentrations in both the topsoil and subsoil are generally highest under Douglas-fir and about equal under Oak and Scots pine. Mg concentrations in the soil solution of the topsoil (Figure 4.4F) are about the same as in the subsoil (Figure 4.5F). Mg concentrations in both the topsoil and subsoil are generally highest under Douglas-fir and lowest under Scots pine.

Changes in average ratios of Al/Ca, Al/(Ca+Mg+K),  $NH_4/NO_3$  and  $NH_4/Mg$  in the topsoil over the years 1990 – 2001 are shown in Figure 4.6.

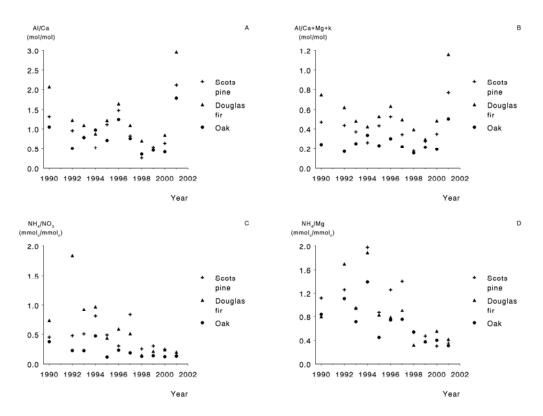


Figure 4.6 Average nutrient ratio's of Al/Ca (A), Al/Ca+Mg+K (B),  $NH_4/NO_3$  (C) and  $NH_4/Mg$  (D) in the topsoil (0-30 cm) for the tree species Douglas Fir, Scots pine and Oak over the yeas 1990 – 2001.

There are no clear trends in these ratios. The Al/Ca ratio is generally above an initially assumed critical value of 1, but more recently a critical value near 1 (Ulrich, 1983) is mostly used for the Al/(Ca+Mg+K) ratio, and this value is hardly ever

exceeded. The  $NH_4/Mg$  ratio is always far below an assumed critical value of 5 (Boxman et al., 1988), due to high nitrification, indicated by the relatively low  $NH_4/NO_3$  ratio (mostly below 0.5).

#### 4.3.2 Trends in chemical composition of the soil solution

To explore if there is a significant trend in soil solution concentrations over time, several linear-regression analyses were performed with year, plot and tree species as explanatory variables. The concentrations in the soil solution in the topsoil (0 - 30 cm) and subsoil (30-60 cm) were used as dependant variables while, at first, year and tree species were used as explanatory variables, according to:

$$Y-concentration = a_0 + a_1 \text{ year} + a_2 \text{ tree species}$$
(4.5)

where Y-concentration is the concentration of element Y in  $mmol_c.l^{-1}$  and year is calendar year AC. Regression coefficients, standard errors and the number of significant coefficients (a<sub>2</sub>) for different tree species are presented in Table 4.9. The SO<sub>4</sub>, Al, K and Mg concentrations in the soil solution of the topsoil and subsoil show a significant overall decrease over the period 1990-2001. For NH<sub>4</sub>, there is a significant decrease in the topsoil and for NO<sub>3</sub> in the subsoil. The SO<sub>4</sub> concentration levels differ significantly under all 3 tree species, whereas all other elements have significant different concentration levels for 2 out of 3 tree species (except for K in the subsoil where only one tree species differs significantly). The decrease of SO<sub>4</sub> and NO<sub>3</sub> concentrations can be explained by the decrease in deposition.

Depth	Substance	$a_0$	a <sub>1</sub>	se a <sub>1</sub>	$\mathbb{R}^2$	nr of tree species with a <sub>2</sub> significant
0-30	SO <sub>4</sub>	177	-0.088	0.014	33	3
	$NO_3$	-11	0.0056	0.011	10	3
	$\rm NH_4$	42	-0.021	0.0038	25	2
	Al	48	-0.024	0.010	31	2
	Κ	26	-0.013	0.0029	25	2
	Ca	-6	0.0032	0.0049	16	2
	Mg	12	-0.0059	0.0028	28	2
60-100	$SO_4$	159	-0.079	0.008	55	3
	$NO_3$	43	-0.021	0.009	13	2
	$\rm NH_4$	6	-0.0028	0.0016	4	2
	Al	97	-0.049	0.010	43	2
	Κ	11	-0.0052	0.0019	6	1
	Ca	-15	0.0075	0.0050	19	2
	Mg	12	-0.0059	0.0018	39	2

 Table 4.9
 Model characteristics and significance of predicting variables for models explaining the concentrations in the soil solution over the years related to tree species

The decreasing Al, K and Mg concentration can be explained by less buffering due to the decrease in potential acid deposition. Ca concentrations in the subsoil show no significant change. In the topsoil, the changes are generally lower due to the large impact of the nutrient cycle (litter fall, mineralization and root uptake), in addition to atmospheric deposition, on the soil solution chemistry of the topsoil.

Secondly separate regression models were fitted for each of the tree species according to:

 $Y-concentration = a_0 + a_1 \text{ year} + a_2 \text{ location}$ (4.6)

Regression coefficients, standard errors and the number of significant coefficients  $(a_2)$  for different locations are presented in Table 4.10. The SO<sub>4</sub>, NH<sub>4</sub> and K concentrations in the soil solution of the topsoil show a significant decrease for each of the tree species over the period 1990-2001 but the decrease differs substantially between tree species and substances. Oak shows the lowest decrease in SO<sub>4</sub>, NH<sub>4</sub> concentration but the largest decrease in K concentrations. In the soil solution of the subsoil only the decrease in SO<sub>4</sub> and Mg concentrations is significant for each of the three tree species. Furthermore, there is a significant decrease in the Al concentration in the subsoil below Scots pine and Douglas-fir. The highest decrease in SO<sub>4</sub> concentrations in the soil solution of both the topsoil and subsoil occurs under Douglas.

# 4.4 Relationships between crown condition, chemical composition of foliage and soil solution with atmospheric deposition

Trends in deposition, chemical composition of the soil solution and foliar chemistry show a decrease in sulphur and nitrogen suggesting a relationship. To analyze if a relation between deposition, foliar chemistry and chemistry of the soil solution exists cross correlation tables were made. For the locations Dwingeloo, Hardenberg, Speuld and Zeist correlations are calculated using paired observations for similar locations and years. Because measurement frequencies are not always similar, the number of paired observations is often smaller than for observations used to analyze temporal trends, thus reducing the number of correlations that can be calculated. Correlations between element deposition and defoliation were also investigated but these appeared to be very low (highest correlation of 0.26 between defoliation and S deposition) which followed already from the different trends in crown condition and atmospheric deposition, as discussed before. In Table 4.11, correlations between nutrient deposition and nutrient content in the foliage are presented for each tree species.

Looking at correlations between similar nutrients in deposition and foliage, most striking is the difference between tree species; with correlations between the deposition and foliar content of S and Mg which are very strong for Oak and Pine but low for Douglas. Pine also shows strong correlation for N in deposition and foliage. For S deposition the strong positive correlation with N foliar content for Oak and the strong negative correlation with Ca foliar content for Oak are salient.

Layer	Element	Tree	$a_0$	a <sub>1</sub>	se a <sub>1</sub>	$\mathbb{R}^2$	nr of plots with a
		species					significant
0-30	SO <sub>4</sub>	Pine	193	-0.09637	0.01177	60	2/5
		Douglas	317.8	-0.1589	0.0392	24	2/5
		Oak	89.33	-0.04463	0.008919	36	2/5
	$NO_3$	Pine	55.92	-0.02783	0.01305	11	1
		Douglas	-58.16	0.02938	0.01857	55	3
		Oak	10.99	-0.00525	0.01551	*	1
	$\rm NH_4$	Pine	40.74	-0.02034	0.006316	20	1
		Douglas	65.76	-0.03286	0.007842	44	2
		Oak	17.11	-0.00847	0.003045	36	4
	Al	Pine	73.91	-0.03679	0.009112	36	2
		Douglas	54.96	-0.02728	0.02322	29	3
		Oak	8.843	-0.00428	0.006405	15	1
	Κ	Pine	19.16	-0.00955	0.002984	16	1
		Douglas	32.54	-0.01624	0.00617	18	1
		Oak	37.4	-0.01868	0.005723	21	2
	Ca	Pine	32.85	-0.0163	0.007714	22	2
		Douglas	-19.78	0.01013	0.008411	25	2
		Oak	-23.54	0.01193	0.007775	15	2
	Mg	Pine	23.28	-0.01158	0.002739	31	2
	0	Douglas	15.71	-0.00776	0.006413	28	3
		Oak	6.004	-0.00294	0.003733	*	1
60-100	SO <sub>4</sub>	Pine	153.9	-0.07691	0.01058	63	3
		Douglas	243.3	-0.1214	0.01824	49	1
		Oak	96.31	-0.04808	0.01159	24	1
	$NO_3$	Pine	65.06	-0.03238	0.01082	36	2
		Douglas	62.3	-0.03101	0.0159	56	5
		Oak	37.84	-0.01872	0.01249	8	1
	$\rm NH_4$	Pine	7.717	-0.00383	0.002449	1	1
	,	Douglas	1.831	-0.00086	0.003276	18	2
		Oak	8.745	-0.00433	0.002807	16	1
	Al	Pine	113.6	-0.05679	0.01121	52	2
		Douglas	171.3	-0.08544	0.02247	20	1
		Oak	2.614	-0.00121	0.007076	73	2
	Κ	Pine	8.073	-0.00402	0.002214	17	2
		Douglas	13.92	-0.00693	0.00461	2	1
		Oak	20.99	-0.01049	0.003029	30	2
	Ca	Pine	16.57	-0.00815	0.006844	10	1
	0	Douglas	-48.07	0.02434	0.01139	17	1
		Oak	-9.655	0.004997	0.00608	11	1
	Mg	Pine	17.86	-0.00887	0.001555	53	2
	-''8	Douglas	16.68	-0.00825	0.001333	51	4
		Oak	16.67	-0.00829	0.002317	31	2

Table 4.10Model characteristics and significance of predicting variables of models for each tree speciesexplaining the concentrations in the soil solution over the years related to plot locations

In Table 4.12 correlations between nutrient deposition and concentrations in the soil solution are presented for two depths.

Tree	Element	S <sub>dep</sub>	$N_{dep}$	K <sub>dep</sub>	Ca <sub>dep</sub>	Mg <sub>dep</sub>
species						
Pine	$ctS_{\rm fol}$	0.951	0.422	0.422	-0.022	-0.633
	$\mathrm{ctN}_{\mathrm{fol}}$	0.289	0.89	-0.626	-0.714	0.324
	$ctK_{fol}$	0.007	-0.434	0.651	0.231	-0.345
	$ctCa_{fol}$	0.079	-0.45	0.736	0.448	-0.495
	$\mathrm{ctMg_{fol}}$	-0.695	0.405	-0.926	-0.822	0.968
Douglas	$ctS_{fol}$	0.344	0.173	0.219	0.014	0.201
	$ctN_{\rm fol}$	-0.232	-0.088	0.371	0.471	0.389
	$ctK_{fol}$	-0.198	-0.037	-0.147	-0.270	0.259
	$ctCa_{fol}$	0.166	0.320	-0.376	-0.644	0.162
	$\mathrm{ctMg_{fol}}$	0.106	0.026	0.182	-0.185	0.320
Oak	$ctS_{fol}$	0.952	0.732	-0.709	0.363	0.074
	$ctN_{\rm fol}$	0.809	0.333	-0.147	0.858	0.681
	$ctK_{fol}$	-0.591	-0.599	0.994	0.165	0.448
	$ctCa_{fol}$	-0.936	-0.72	0.739	-0.342	-0.049
	$\mathrm{ctMg_{fol}}$	-0.147	-0.537	0.826	0.767	0.922

Table 4.11 Correlations between nutrient deposition and foliar chemistry

Table 4.12 Correlations between nutrient deposition and chemistry of the soil solution

Layer	Element	Deposi	tion						
		SO <sub>4</sub>	NO <sub>3</sub>	$\rm NH_4$	Κ	Ca	Mg	Na	Cl
0-30	$[SO_4]$	0.14	-0.053	-0.101	0.831	0.15	0.219	-0.135	-0.192
	$[NO_3]$	0.463	0.254	0.254	0.066	-0.176	-0.087	-0.203	-0.344
	$[NH_4]$	0.017	0.388	0.174	0.428	0.255	0.293	-0.071	-0.14
	[Al]	0.145	0.251	0.078	0.592	0.315	0.229	-0.305	-0.365
	[K]	-0.147	-0.651	-0.528	0.176	-0.332	-0.338	-0.406	-0.404
	[Ca]	0.357	0.367	0.051	0.398	0.201	0.398	0.285	0.07
	[Mg]	0.084	0.342	0.121	0.523	0.193	0.303	-0.051	-0.16
	[Na]	-0.041	0.258	-0.004	0.81	0.383	0.541	0.201	0.095
	[Cl]	-0.15	0.28	0.001	0.652	0.416	0.492	0.104	0.024
60-100	$[SO_4]$	0.497	-0.041	0.123	0.22	-0.186	-0.18	-0.282	-0.293
	$[NO_3]$	0.628	0.288	0.253	0.358	0.126	0.171	-0.195	-0.27
	$[NH_4]$	0.367	0.51	0.409	0.189	0.201	0.218	-0.146	-0.173
	[Al]	0.572	0.111	0.247	-0.182	-0.209	-0.13	0.051	0.051
	[K]	0.239	0.164	0.008	-0.327	-0.334	-0.03	0.394	0.287
	[Ca]	0.562	0.122	0.146	0.06	-0.266	-0.054	0.033	-0.081
	[Mg]	0.515	0.449	0.352	0.131	0.044	0.185	-0.083	-0.127
	[Na]	0.269	0.699	0.427	-0.115	0.226	0.457	0.437	0.338
	[Cl]	0.021	0.541	0.126	0.369	0.479	0.762	0.462	0.355

Most striking is the absence of strong correlations between similar nutrients in deposition and soil solution, with the exception of  $SO_4$  and  $NH_4$  in the subsoil. Correlations between nutrient deposition and foliar chemistry are much stronger indicating a direct link between deposition and foliar chemistry. The occurrence of stronger correlations for the soil solution deeper in the soil profile is due to the previously mentioned larger impact of the nutrient cycle, in addition to atmospheric deposition, on the soil solution chemistry of the topsoil.

# 5 Changes in growth and species composition of the ground vegetation

# 5.1 Growth changes over the period 1995 – 2005

#### Data analyses

Information on periodic annual increment at the Intensive Monitoring plots has become available since 1995 because of two re-measurements of the trees, five years (2000) and ten years (2005) afterwards. The repeated data on tree diameter (at breast height) and tree height were used to calculate standing wood volume and changes therein. In presenting information on the volume and volume changes of wood in forests, it is important to define the type of wood included, such as all stem wood from the bottom, stem wood above a minimum diameter, total above-ground woody biomass etc. (De Vries et al., 2003). In this chapter we report all stem wood from the bottom, as a basis the related increment and carbon pool changes in stem wood during a 10 year period. In the future, an in-depth analysis on the deviation in expected growth (based on standard growth curves for the plot) and the natural and anthropogenic growing conditions, such as stand and site characteristics, soil chemical variables, meteorology and atmospheric deposition, is worthwhile to be carried out.

Based on the measured diameter at breast height (dbh) and tree height (h), the volume (dm<sup>3</sup> or l) of each tree was calculated according to the following relationships (Dik et al., 1996):

$$Volume = \exp(a_1 \cdot \ln(dbh) + a_2 \cdot \ln(h) - a_3)$$
(5.1)

where dbh is diameter at breast height in cm and h is tree height in m. Values used for  $a_1$ ,  $a_2$  and  $a_3$  are given in Table 5.1. To estimate changes in stem wood volume total carbon pools for each plot, the dbh and height of all trees in a plot need to be measured for all three years, but this appeared not to be the case for height. The absence of measurements trees was corrected by using a factor, as described in Section 2.2.6.

Tree species (	Broup	a <sub>1</sub> ,	a <sub>2</sub>	a3
Pine	Pinus sylvestris	1.93255	0.85915	2.62597
Douglas Fir	Pseudotsuga menziesii	1.83654	0.93879	2.61466
Oak	Quercus Robur	1.86115	1.039	2.95925

 Table 5.1
 Coefficients used to calculate stem wood volumes in dm<sup>3</sup> (or l) per tree species.

By multiplying single tree volume with wood densities and tree carbon contents, an estimate for the carbon pool stored in the stem was derived and extrapolated to carbon pools per hectare. Values used for wood densities are given in Table 5.2. For the carbon content of wood we used an estimated value of 50%.

Table 5.2 Stem wood densities per tree species that were used to calculate carbon pools in trees

Tree species		Wood density (kg.m <sup>-3</sup> )
Pine (SP)	Pinus sylvestris	490
Douglas Fir (DF)	Pseudotsuga menziesii	470
Oak (OA)	Quercus Robur	600

Because tree measurements were only conducted in 1995, 2000 and for some plots in 2005, a temporal trend was not calculated. Instead differences in carbon pools of standing wood were calculated between 1995 and 2000 and between 2000 and 2005 for all the trees in a plot. For the plot in Schoonloo, tree height was not measured in 2000 but an average height was estimated for nearly all trees together, therefore volumes of individual trees and the total volume could not be calculated.

#### Results

The stem wood volume and the related carbon pools in stem wood for the various locations are presented in Table 5.3 and graphically depicted in Figure 5.1 and Figure 5.2. To allow a comparison of changes in stem wood volume (Table 5.3 and Figure 5.1) and related carbon pools (Table 5.3 and Figure 5.2), the amounts are given per ha for each level 2 plot.

Plot	Tree <sup>1)</sup>	Volume	(m <sup>3</sup> /ha)		C-pool (	ton/ha)	
		1995	2000	2005	1995	2000	2005
Gasselte	DF	271	286		64	68	
Smilde-1	SP	177	243		43	59	
Smilde-2	SP	177	211		44	53	
Schoonloo	DF	120			28		
Hardenberg	DF	333	495	543	79	118	129
Kootwijk-2	DF	340	458		80	108	
Leende-1	SP	163	237		40	58	
Leende-2	SP	208	232	258	51	57	63
Ulvenhout	OA	151	183		45	55	
Speuld-1	OA	159	205		48	61	
Zeist	OA	133	173	186	38	49	53
Hoenderloo	OA	196	233		57	68	
Speuld-2	DF	440	537	750	103	126	176
Dwingeloo	SP	155	218	231	38	54	57

Table 5.3 Stem wood volume and carbon pools in stem wood in 1995, 2000 and 2005 of the level 2 sites

SP = Scots pine (Pinus sylvestris), DF= Douglas fir (Pseudotsuga menziesii) and OA =Oak (Quercus robur)

Large stem wood volumes and C-pools can be found at Speuld-2, Kootwijk-2 and Hardenberg, where the dominant tree species is Douglas-fir. These three locations also show the highest growth rates; on average 25 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup> in the first period. The dominant tree species at the plot in Schoonloo is also Douglas-fir but the stem volume and C-pool are smallest of all plots here. Except this plot, the stem volumes of Douglas fir sites are above 250 m<sup>3</sup>.ha<sup>-1</sup>. Plots were the dominant tree species is Scots pine on average show intermediate stem volumes (176 m<sup>3</sup>.ha<sup>-1</sup>) and growth rates (10 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup>). Plots were the dominant tree species is Oak on average show the smallest stem volumes (160 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup>) and growth rates (8 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup>). The

carbon pool changes are generally 4 times as low as the volume changes, being near 6 ton C.ha<sup>-1</sup>.yr<sup>-1</sup> for Douglas fir and near 2.5 ton C.ha<sup>-1</sup>.yr<sup>-1</sup> for both Pine and Oak. If recorded, the volume increase between 2000 and 2005 is generally small compared to the increase between 1995 and 2000, except for Speuld-2 where growth is about twice as high in the second period. The situation at Speuld-2 is exceptional because growth rates normally decline with age of the forest.

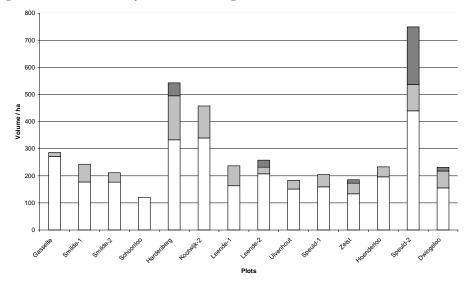


Figure 5.1 Stem volumes  $(m^3/ha)$  in 1995 (white), 2000 (white and gray) and 2005 (white and gray and black) for each level 2 plot

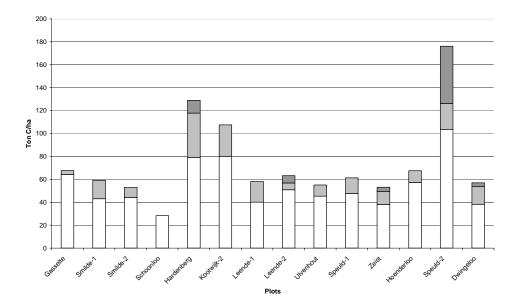


Figure 5.2 Carbon pool (ton C/ha) in 1995 (white), 2000 (white and gray) and 2005 (white and gray and black) for each level 2 plot.

## 5.2 Changes in species composition over the period 2000 – 2005

## Data analyses

Changes in ground vegetation composition can be used to estimate changes in abiotic variables that influence the vegetation. If the changes in abiotic variables inferred from the vegetation changes agree with measured abiotic changes one can be confident that (a) the assumed determinative variables really affect vegetation, and (b) the changes in these variables are also reflected in the vegetation. For instance: deposition negatively affects vegetation, and a reduction in deposition leads to an increase in biodiversity. Before determining the magnitude, direction and possible causes of vegetation changes, the variation in the vegetation at a single point in time (i.e. 14 plots in 2000) was evaluated first. We then evaluated the change between 2000 and 2005, but this analysis was strongly limited due to the scarcity of data, since the re-sampling in 2005 only took place at 5 plots.

The evaluation of vegetation changes in the light of abiotic changes can be done in a two-step procedure, namely (1) determination of the effect of measured abiotic variables on the vegetation by regression, and (2) use of the regression parameters and the observed vegetation change to infer abiotic changes. However, both steps can be combined into a single procedure as described by Van Dobben and Ter Braak (1998) and Van Dobben and Slim (2005). In the absence of abiotic measurements, expert judgement of the response of the vegetation to its environment can be used to qualitatively infer abiotic changes.

The present analysis focuses on (1) the variation in the vegetation of the 14 plots in 2000, and (2) the change in the vegetation between 2000 and 2005 of the 5 plots that were surveyed at both dates. Both univariate and multivariate techniques were applied, using the computer programs (Payne et al., 2003) and CANOCO (Ter Braak & Šmilauer, 2002) respectively.

Before analysis, the cover codes were transformed to percentages according to Table 2.15. Subsequently, the cover percentages were averaged over the four subplots, and these were treated as the basic observations. The averaged cover percentages were LN(X+1) transformed before the statistical analysis in order to approximate normality. The moss, herb and shrub layers were combined to the maximum cover per species. The tree layer was excluded from the vegetation and used as an explanatory variable instead.

## Results

## Variation in 2000

The surveyed plots are poor in species. In 2000, a total of 57 vascular and 37 cryptogamic species was found. Table 5.4 give the most common vascular and cryptogamic species, respectively.

Name	Code	Numbers found
Vascular species		
Pseudotsuga menziesii	Pseutmen	76
Quercus robur	Quercrob	63
Pinus sylvestris	Pinussyl	52
Deschampsia flexuosa	Deschfle	52
Rhamnus frangula	Rhamnfra	50
Dryopteris carthusiana	Dryopcar	48
Sorbus aucuparia	Sorbuauc	47
Prunus serotina	Prunuser	46
Dryopteris dilatata	Dryopdil	42
Fagus sylvatica	Fagussyl	30
Ceratocapnos claviculata	Ceraccla	23
Quercus rubra	Quercrub	17
Betula pendula	Betulpen	17
Amelanchier lamarckii	Amelalam	16
Carex pilulifera	Carexpil	14
Vaccinium myrtillus	Vaccimyr	12
Picea abies	Piceaabi	12
Empetrum nigrum	Empetnig	11
Molinia caerulea	Molincae	10
Agrostis capillaris	Agroscap	9
Calluna vulgaris	Calluvul	9
Ilex aquifolium	Ilex Aqu	9
Galium saxatile	Galiusax	8
Galeopsis tetrahit	Galeotet	7
Chamerion angustifolium	Chameang	6
Lonicera periclymenum	Lonicper	6
Cryptogamic species		
Hypnum jutlandicum	hypnujut	68
Brachythecium rutabulum	bractrut	51
Eurhynchium praelongum	eurhypra	48
Dicranum scoparium	dcnumsco	46
Lophocolea heterophylla	lophchet	44
Campylopus pyriformis	campspyr	43
Dicranella heteromalla	dcllahet	43
Pleurozium schreberi	plrozsch	39
Polytrichum formosum	polymfor	35
Lophocolea bidentata	lophcbid	26
Mnium hornum	mniumhor	23
Pseudoscleropodium purum	pseucpur	20
Orthodontium lineare	ortholin	19
Aulacomnium androgynum	aulacand	18
Plagiothecium undulatum	plagtund	17
Campylopus flexuosus	campsfle	16
Hypnum cupressiforme	hypnucup	13
Plagiothecium denticulatum	plagtden	12
Campylopus introflexus	campsint	11
Dicranoweisia cirrata	dcnowcir	7
Pohlia nutans	pohlinut	6
1 011114 millions	Polimicat	0

Table 5.4 List of vascular and cryptogamic species that were found more than 5 times.

Multivariate techniques were used to form a general picture of the variation in the vegetation. The species turnover appeared to be low (gradient length is ca. 3 SD units) which justifies the use of linear methods (PCA and RDA). Using the tree species as explanatory variable in RDA, only Scots Pine (*Pinus sylvestris*) and Douglas Fir (*Pseudotsuga menziesii*) had a significant effect. The effect of Oak *Quercus robur* is not significant, and the other tree species have too few occurrences to be used as explanatory variables (Table 5.5).

Table 5.5 Summary of RDA analysis of the effect of tree species on ground vegetation. AX1 etc.: RDA axes; <sup>1</sup> eigenvalue \* 100%.

Variable	Р	F	%ExplVar	%ExplVar
				(Cum)
Pinus Syl <sup>1</sup>	0.002	3.56	23%	23%
Pseutmen <sup>2</sup>	0.027	2.11	12%	35%
Quercrob <sup>3</sup>	0.446	1.02	6%	41%
AX1			24%1	24%
AX2			13%1	37%
AX3			5%1	41%
AX4			19%1	-

<sup>1</sup> Pinus Syl = *Pinus sylvestris* or Scots Pine.

<sup>2</sup> Pseutmen<sup>2</sup> = *Pseudotsuga menziesii* or Douglas Fir.

<sup>3</sup> Quercrob<sup>3</sup> = *Quercus robur* or Oak.

Figure 5.3 shows the PCA biplot based on all data from 2000, with the tree species added as 'indirect' explanatory variables (i.e. the position of the understorey species is solely determined by their mutual correlation and not by their correlation with the tree species). Figure 5.3 shows that a high cover of *Pinus* coincides with species than commonly occur in heath land (e.g., *Calluna vulgaris, Desschampsia flexuosa, Empetrum nigrum*) while a high cover of *Pseudotsuga* coincides with moss species (e.g. *Eurhynchium spp., Polytrichum formosum, Mnium hornum*).

Figure 5.4 shows the Ellenberg indicator values for light (A) and nutrients (B) projected into the plot of Figure 5.3. Figure 5.4A suggests that light intensity might be a causal factor behind these differences, however the correlation between the scores on the first two axes and the Ellenberg indicator for light was not significant (data not shown). Also the correlation between the scores on the first two axes and the Ellenberg indicator for nutrient availability was not significant and the differences in nutrient availability seem to be small (Figure 5.4B).

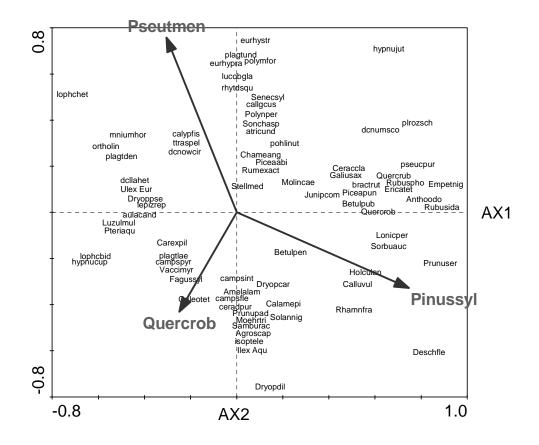


Figure 5.3 PCA biplot of species and 'environmental' variables (= the three most common trees species) as indirect explanatory variables. Explanation of abbreviated species names in Tables \* and \*. Note that vascular species codes start with Uppercase, cryptogam codes start with lowercase.

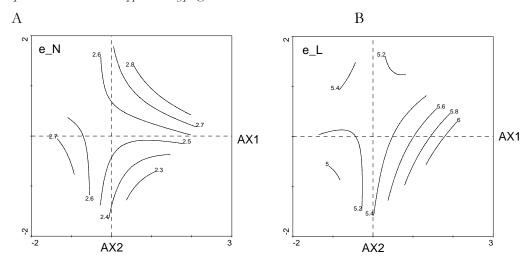


Figure 5.4 Mean Ellenberg indicator values projected into the plot of Figure 5.3. Smoothing by 2nd-order polynomial. Left: light indicator, right: nutrient indicator. Ellenberg values are unweighted mean values of all species present in a plot.

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#### Change over time

The change over time was determined for only the five plots that were sampled twice, i.e. in 2000 and 2005. To do this, both multivariate and univariate (species by species) techniques were used. The multivariate technique was an RDA, which was performed with the year number as the only explanatory variable. Its effect appeared to be non-significant (P=0.9), even though the plots themselves were not included as co-variables. Figure 5.5 is the biplot of Figure 5.3 with the observations from 2005 added as 'passive' observations.

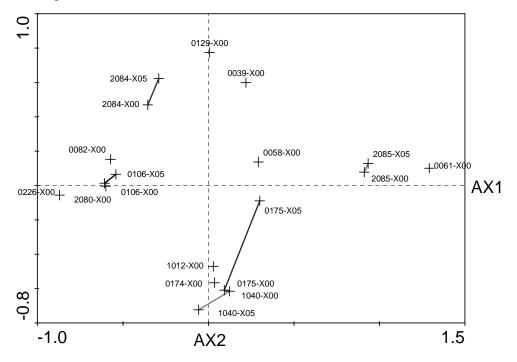


Figure 5.5 Change in sample scores projected into the plot of Figure 5.3. Scores of each single plot at both points in time are connected by lines (X00: in 2000; X05: in 2005). Plots in 2005 are 'passive' samples i.e. the axes are determined by the observations in 2000. Plots without a line have only one observation (X00: in 2000).

The results of the univariate approach is given in Table 5.6, which presents the difference (2005 minus 2000) in the untransformed percentage cover for the species that were found in at least two plots in at least one point in time, and have more than five occurrences (included in Table 5.4). In general, the changes have been small. Only two species increased by more than 10% cover (the mosses *Hypnum jutlandicum* and *Pleurozium schreberi*) and two decreased by more than 10% cover (the moss *Dicranum scoparium* and the grass *Desschampsia flexuosa*). The change was statistically significant for only four species, interestingly not the ones with the largest absolute change (the tree seedlings *Quercus rubra* and *Picea abies*, and the moss *Aulacomnium androgynum* decreased significantly, and the moss *Atrichum undulatum* increased significantly). The number of increasing species per plot decreased from 21 to 15.

Change over time				
Species	Mean Diff	Т	Significance	Number Plots
Deschampsia flexuosa	-23.79	-0.82		4
Dicranum scoparium	-7.47	-0.50		4
Ceratocapnos claviculata	-1.10	-0.68		3
Lophocolea heterophylla	-0.21	-1.22		4
Sorbus aucuparia	-0.18	-0.49		4
Dicranella heteromalla	-0.12	-0.96		3
Quercus robur	-0.09	-0.79		3
Calluna vulgaris	-0.07	-1.31		2
Lophocolea bidentata	-0.05	-2.00		5
Carex pilulifera	-0.05	-0.42		2
Quercus rubra	-0.05	-4.50	*	4
Picea abies	-0.04	-2.83	*	2
Aulacomnium androgynum	-0.04	-2.83	*	2
Campylopus flexuosus	-0.03	-0.61		4
Orthodontium lineare	-0.02	-0.71		2
Amelanchier lamarckii	-0.02	-0.71		2
Juniperus communis	-0.02	-0.71		2
Campylopus pyriformis	-0.01	-0.19		5
Molinia caerulea	-0.01	-0.14		2
Agrostis capillaris	0.02	0.71		2
Ilex aquifolium	0.02	1.10		5
Hypnum cupressiforme s.l.	0.07	0.28		2
Brachythecium rutabulum	0.12	0.12		5
Pseudoscleropodium purum	0.46	0.69		4
Pinus sylvestris	0.52	0.57		3
Atrichum undulatum	0.74	34.65	*	2
Dryopteris dilatata	0.84	0.66		5
Dryopteris carthusiana	0.91	0.50		5
Mnium hornum	1.52	0.46		2
Eurhynchium praelongum	2.47	0.70		5
Pseudotsuga menziesii	2.57	0.32		3
Polytrichum formosum	2.59	0.64		3
Rubus fruticosus	3.71	0.71		2
Prunus serotina	3.85	0.99		4
Rhamnus frangula	4.70	0.84		4
Fagus sylvatica	4.98	0.70		2
Hypnum jutlandicum	7.69	0.65		5
Pleurozium schreberi	17.06	0.57		4

Table 5.6 change between 2000 and 2005 as difference in untransformed percentage cover (2005 minus 2000), and statistical significance. T = T-values of paired Student's T-test; Number Plots = number of plots with this species in at least one point in time. Species are in order of increasing change.

The change might be interpreted as a tendency towards an understorey vegetation of darker forest (increase of mosses, decrease of 'heathland species' like *Deschampsia flexuosa, Calluna vulgaris, Carex pilulifera*). This pattern is corroborated by the generally upward displacement of the sample scores in the multivariate analysis of Figure 5.5. The decrease of *Deschampsia flexuosa* agrees with the idea that dominance of this species is typical for the succession stage from which Dutch forests are now moving away. In a next stage *Deschampsia flexuosa* should be replaced by *Vaccinium myrtillus.*, however this species was present in only one plot where it changed only very slightly.

## Conclusions

On the basis of this analysis, the following conclusions can be drawn

- the plots are poor in species
- differences in species composition in the ground vegetation in 2000 can largely be explained from the difference in dominant tree species
- species composition indicates acid and nutrient poor conditions
- these conclusions could be further elaborated with the aid of soil chemical data
- the changes between 2000 and 2005 were small, and the small number of plots hardy allowed the detection of a pattern in the change
- no abiotic changes are evident from the vegetation changes

## 6 Water fluxes and nutrient budgets in the period 2003-2005

## 6.1 Introduction

A comparison of element inputs from the atmosphere and element outputs from the bottom of the root zone give insight in the fate (accumulation or release) of sulphur (S), nitrogen (N), base cations (BC) and aluminium (Al) in the ecosystem, where base cations stands for the sum of calcium (Ca), magnesium (Mg) and potassium (K). More specifically, budgets of SO<sub>4</sub>, NO<sub>3</sub> and NH<sub>4</sub> give insight in (i) the actual rate of acidification due to anthropogenic sources and (ii) the potential rate of acidification by immobilisation of S and N (e.g. Van Breemen et al., 1984; De Vries et al., 1995). Results on the input and output of Al and BC give information about the mechanisms buffering the acid input (e.g. Mulder & Stein, 1994; Wesselink, 1994; De Vries et al., 1995). In general, the ratio of Al to BC release is a crucial aspect with respect to soil mediated effects of acid inputs (e.g. Cronan et al., 1989; Sverdrup & Warfvinge, 1993). These insights can therefore be used to derive critical deposition levels for forest soils (ecosystems). Comparison with available data on present loads, leads to insight in the stress of air pollution on the chemical ecosystem condition (e.g. De Vries et al., 2000). As such, it is of crucial importance to assess the present and future impacts of atmospheric deposition on the element cycle and nutrient availability.

Element budgets have already been carried out at Intensive Monitoring plots by several countries including Ireland (Boyle et al., 2000; Farrell et al., 2001), Germany (Sprangenberg, 1997; Wetzel, 1998; Block et al., 1999) and Slovakia (FIMCI, 2000). Furthermore, there are several literature compilations of element budgets, focusing on the behaviour of N (e.g. Dise et al., 1998a; Dise et al., 1998b; Gundersen et al., 1998b; MacDonald et al., 2002), base cations (Armbruster et al., 2002) and Al (Dise et al., 2001). A European wide assessment of element budgets, using all available data on deposition, meteorology and soil solution chemistry at Intensive Monitoring plots has recently been published as well (De Vries et al., 2007). In that paper, no information was included on Dutch plots, because the results of the lysimeter monitoring carried out since 2003 were not yet available.

This section aims to fill this gap. It describes the methods used to calculate output and retention or release of major elements (specifically focused on  $SO_4$ , N, BC and Al) and gives results on the temporal variation in the years 2003-2005 for three plots (Zeist, Hardenberg and Dwingeloo). For the location of Hardenberg (plot 106), Zeist (plot 1040) and Dwingeloo (plot 2085), we refer to Section 2.1. Special features of the sites, specifically related to the average groundwater level, which is relevant for the assessment of water fluxes and related nutrient fluxes, are:

- Hardenberg: The average groundwater level is near 1.2 meter below surface. There is a ditch with a depth of two meter along the site which influenced the hydrology of the site strongly.
- Zeist: The average groundwater level is more than 4 meter below surface.

- Dwingeloo: The average groundwater level is near 2 meter below surface (measured at the lowest location within the site), but there are large differences caused by large (about 6 meters) differences in height on the site caused by former sand dunes. Furthermore, the site contains a thick layer of heath and mosses of approximately 30 cm, largely influencing the calibrated stand characteristics of the site.

## 6.2 Methods

## 6.2.1 Water fluxes

Hydrological fluxes at the plots Hardenberg, Zeist and Dwingeloo where calculated with the soil hydrological model SWAP. SWAP (Soil, Water, Atmosphere and Plant) simulates transport of water, solutes and heat in unsaturated/saturated soils. For a detailed description of the SWAP model see the Reference Manual SWAP version 3.0.3 (Kroes & van Dam, 2003) and the PhD-thesis of Van Dam (Van Dam, 2000).

To calculate the hydrological fluxes the model SWAP needs: (i) meteorological data, (ii) abiotic characteristics of the site (soil physical characteristics, drainage characteristics) and (iii) forest stand characteristics (such as: crop resistance, leaf area index (LAI), soil cover, storage capacity of the crown, tree height and root distribution). A short overview of the data thus derived is given below.

## Meteorological data

The SWAP model uses daily data on global radiation, minimum and maximum temperature, relative humidity, wind speed, precipitation and reference evaporation. The daily global radiation was calculated using the FAO (FAO, 1998) method based on relative sunshine hours and latitude of the site. The reference evaporation was calculated with the method of Makkink (Makkink, 1957), using the calculated global radiation. Maximum, minimum temperature, wind speed and relative humidity where taken from a nearby meteorological head station of the National Dutch Weather station KNMI. Daily precipitation (bulk deposition) was taken from a nearby precipitation station of KNMI (see Table 6.1). At Hardenberg (plot 106), monthly bulk deposition was measured by ECN, whereas bulk deposition for the plots 2085 Dwingeloo and 1040 Zeist were taken from the nearest RIVM deposition stations (see also Section 2.2.1).

Precipitation may vary considerably over short distances, potentially leading to large differences in daily precipitation between data from a (nearby) meteorological station and the actual measured precipitation at the site. To obtain a best estimate of the daily precipitation at the site, daily precipitation data has been corrected on the measured precipitation (bulk deposition) at the open field close to sites according to:

$$P_{i,site} = P_{i,station} \cdot \frac{P_{period,site}}{P_{period,station}}$$
(6.1)

where  $P_{i,site}$  is the daily precipitation at the site,  $P_{i,station}$  is the daily precipitation measured at the meteorological precipitations station,  $P_{period,site}$  is the measured precipitation at the site during a period of one month and  $P_{period,station}$  is the precipitation measured at the meteorological precipitation station during this period.

Site	KMNI head station	Distance to site (km)	KNMI precipitation station	Distance to site (km)	RIVM deposition station	Distance to site (km)
Hardenberg	Twente	48	Rheezerveen	2.8		
Zeist	De Bilt	5.6	De Bilt	5.6	De Bilt	5.6
Dwingeloo	Eelde	40	Dwingeloo	8.4	Valthermond	54.6

Table 6.1 Origin of the meteorological data used in the model calculations

#### Soil physical and hydrological characteristics

To calculate fluxes and water contents, SWAP needs information on the soil physical characteristic of the soil, data on the hydrological conditions at the bottom of the soil profile and drainage characteristics. Water retention characteristics and saturated conductivity (see Table 6.2) were selected from the Staring soil series (Wösten et al., 1994). Water retention characteristics for the organic litter layer were based on data from a Dutch Douglas fir site (Tiktak & Bouten, 1994).

At the plot in Zeist, groundwater occurs at a depth of more than 4 meters and free drainage of the soil profile was assumed. At the other sites the groundwater was within the upper 3 m of the soil profile. Here, the fluxes at the bottom of the soil profile were calculated as a function of the groundwater level (Ernst & Feddes, 1979):

$$Q_{\rm bot} = a \, e^{b\phi_{\rm avg}} \tag{6.2}$$

with  $Q_{bot}$  is the flux at the bottom boundary, a (cm d<sup>-1</sup>) and b (cm<sup>-1</sup>) are parameters. Values for a and b were -0.714 and -0.0238 for Hardenberg, and -0.247 and -0.0138 for Dwingeloo. The values for a and b were calibrated on measured groundwater levels and soil moisture contents.

Site  $\Theta_{\underline{sat}}$ Depth (cm) Ν K<sub>sat</sub> L  $\theta_{\text{res}}$ А 0-9\* 800 0.5 0.100 1.25 Hardenberg 0.000 0.018 >9 0.32 0.0597 2.059 223.2 0.01 0.343  $0-9^{*}$ Zeist 0.5 0.000 0.100 1.25 800 0.018 > 90.32 0.01 0.0597 2.059 223.2 0.343 Dwingeloo  $0-13^{*}$ 0.0000.5 0.100 1.25 800 0.018 13-103 0.02 0.38 0.0214 2.07563.9 0.039 103-253 0.01 0.34 0.211 1.564 44.58 -0.522 0.039 >253 0.02 0.38 0.0214 2.07563.88

Table 6.2 Soil hydrological characteristics used in the SWAP calculation for Hardenberg, Zeist and Dwingeloo

 $\theta_{res}$  = residual water content,  $\Theta_{sat}$  = saturated water content,  $\alpha$ , n= shape parameters

 $K_{sat}$ = saturated hydraulic conductivity, l = exponent in hydraulic conductivity function \* Litter layer

Lateral drainage to the ditch at Hardenberg was calculated using data on drainage resistance, spacing, depth and water level of the ditch (Table 6.3). Because the water level of the ditches was assumed to be constant during the year, it was only measured once and was used as input for the model. Drainage resistances were calibrated on measured groundwater levels and soil moisture contents. For Zeist and Dwingeloo no lateral drainage was calculated, since ditches and canals were absent.

Table 6.3 Drainage parameters used in the SWAP calculation for Hardenberg

Parameter	Hardenberg
Drainage resistance (d)	800
Ditch spacing (m)	1000
Depth of the ditches (cm)	-130
Average water level in ditches (cm)	-145

## Forest stand characteristics

The most important forest stand characteristics used by the SWAP model are tree height, canopy resistance, leaf area index (LAI), storage capacity of the canopy (Smax), soil cover (sc) and root distribution, as presented in Table 6.4 and Table 6.5. Only tree height was measured. LAI, Smax and Sc were based on model calibration with measured groundwater levels. Data for the water content, canopy resistance and relative root density were derived from a previous literature study on stand characteristics (Van der Salm et al., 2006). The calibration of the stand characteristics of the Dwingeloo Scots-pine site was strongly influenced by the thick litter/heather layer, which adsorbed most of the precipitation. In the model calculations this litter/heather layer is added to the canopy characteristics of the Dwingeloo site (S<sub>max</sub> and SC). The calibration of the site.

Table 6.4 The most important stand characteristics used in the SWAP calculation for Hardenberg, Zeist and Dwingeloo

Plot	Tree species	Age in	Height	LAI	Canopy	Interception parameters		
		2004	(meter)		resistance	S <sub>max</sub>	Sc	
		(year)						
Hardenberg	Douglas-fire	84	28.1	3.0	85	0.15	0.40	
Zeist	Oak	76	18.2	4.0	50	0.09	0.70	
Dwingeloo	Scots pine	79	18.6	5.0	100	0.25	0.99	

Depth (cm)	Hardenberg	Zeist	Dwingeloo
	Douglas-fire	Oak	Scots pine
0-10	0.15	0.20	0.30
10-20	0.15	0.20	0.20
20-30	0.15	0.20	0.10
30-40	0.15	0.10	0.10
40-50	0.10	0.10	0.05
50-60	0.09	0.05	0.05
60-70	0.06	0.05	0
70-80	0.05	0.05	0
80-90	0.05	0.04	0
90-100	0.05	0.01	0

Table 6.5 The relative root density (%)used in the SWAP calculation for Hardenberg, Zeist and Dwingeloo

## 6.2.2 Nutrient budgets

Leaching fluxes of the considered elements were calculated by multiplying the measured soil solution concentrations (as mentioned in Section 2.2.4) with the water leaching fluxes at the corresponding depth. Element leaching fluxes were calculated at a depth of 15 cm and 80 cm, being the two depths at which the soil solution chemistry was measured by using lysimeters. The deepest lysimeter cup at 80 cm is assumed to represent the leaching fluxes at the bottom of the root zone.

Concentrations in lysimeter cups were measured during 12 periods, but the sampling times are not chosen at fixed moments (e.g. monthly intervals) but related to the precipitation surplus, as described in Section 2.2.4. Water fluxes with SWAP were calculated on a daily basis. Leaching fluxes can be calculated by either multiplying (De Vries et al., 2001; 2007):

- The (average) measured concentration during the period between two subsequent measurements with the accumulated water flux during that period (method 1) or
- The daily interpolated concentrations with the daily water fluxes calculated by SWAP (method 2).

The choice between the first and second method described above depends somewhat on the method and the measurement frequency used to sample the soil solution. When the sampling frequency is high, the collected samples will be representative for the average concentration during the sampling period and method 1 may be preferred. When longer sampling intervals are used, daily interpolation of the measured concentrations between two subsequent measurements may be preferred (method 2). In this study method 2 was thus used because of the absence of lysimeter samples in the dry summer periods. In general differences between both approaches are small (De Vries et al., 2001).

Total element budgets were calculated by subtracting the leaching flux (at 80 cm) from the total deposition flux on the forest, in which total deposition fluxes were derived as described in Section 3.1). Positive budgets indicate that a certain element is retained in the soil, whereas negative budgets indicate a net release of this element from the soil. We also present the canopy interactions (difference between total

deposition and throughfall) and the soil budget, being the difference between throughfall and the leaching at 80 cm. It has to be noted that this difference is not equal to the soil retention since the impact of the nutrient cycle (litter fall, mineralisation and root uptake) is not accounted for. The above ground canopy uptake may lead to a reduced root uptake. When the total element amount in the canopy stays constant, the soil retention equals the total deposition flux on the forest minus the leaching flux at 80 cm.

## 6.3 Results

## 6.3.1 Water fluxes

Soil physical and hydrological characteristics and most forest stand characteristics were calibrated on measured groundwater levels and soil water contents. At the Zeist plot the groundwater level is at a depth of more than 4 meters and for model calculations free drainage of the soil profile was assumed. Calibration of the model with measured throughfall data was not possible due to different measuring periods of the bulk deposition and throughfall data. Results of the calibration are given in figure 6.1 and 6.2.

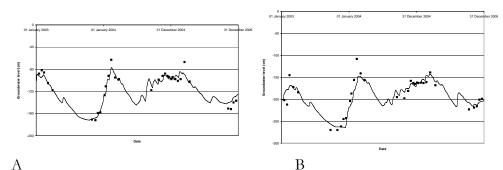
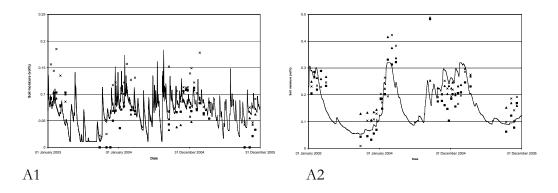


Figure 6.1: Measured (points) and calculated (line) groundwater levels (cm) of the Hardenberg (A), and Dwingeloo (B) site.



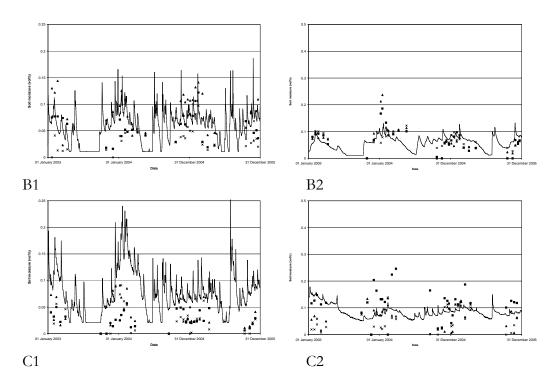


Figure 6.2: Measured (points) and calculated (line) soil moisture contents of the Hardenberg (A), Zeist (B) and Dwingeloo (C) site at 15cm (X1) and 80 cm (X2) depth.

Information on the yearly water fluxes and water balance, in terms of interception evaporation, soil evaporation, transpiration and ultimate ground water recharge by leaching below the root zone (at 80 cm depth) for the period 2003-2005 is given in Table 6.6.

Table 6.6 Average yearly water fluxes and water balances for the period 2003-2005 for Hardenberg, Zeist and Dwingeloo (between brackets the measured values)

Type of flux	Flux (mm/yr)								
	Hardenberg: Douglas-fr	r Zeist: Oak	Dwingeloo: Scots pine						
Precipitation	8091	785 <sup>1</sup>	9211						
Interception	174	130	416						
Throughfall	635 (548)	655 (718)	505 (476)						
Soil evaporation	114	133	45						
Leaching at 15 cm	503	443	324						
Leaching at 80 cm	311	275	135						

<sup>1</sup> Precipitation

The difference between measured precipitation and calculated throughfall equals the calculated interception evaporation. This equals 22% of the precipitation for Hardenberg, 17% for Zeist and 45% for Dwingeloo. If the measured throughfall is used, the measured interception evaporation equals 21, 10 and 35% of the precipitation. The high interception evaporation at Dwingeloo is caused by the thick layer of heath and mosses. For the model calculations this layer is added to the stand characteristics of the Scots pine trees on the plot.

The calculated soil evaporation equals 14, 17 and 5% of the precipitation. The low soil evaporation of the Dwingeloo site is again strongly influenced by the thick heath and moss layer. The difference between the water flux at the soil surface and the leaching flux at 80 cm equals the soil transpiration. This equals 40, 46 and 40% of the precipitation for the Hardenberg, Zest and Dwingeloo site. The precipitation excess decreases going from the Douglas fir at Hardenberg (311 mm.yr<sup>-1</sup>) > Oak at Zeist (275 mm.yr<sup>-1</sup>) > Scots pine at Dwingeloo (135 mm.yr<sup>-1</sup>). This is not typical for these tree species and is due to the site conditions. In general the precipitation excess of these trees decreases going from Oak > Scots pine > Douglas fir.

## 6.3.2 Element budgets

Information on the yearly element fluxes and element budgets, in terms of total deposition, throughfall, leaching below the root zone (at 80 cm depth) and element budgets (total budget = deposition minus leaching at 80 cm depth, soil budget = throughfall minus leaching at 80 cm) for the period 2003-2005 is given in Table 6.7. For Zeist, where the average groundwater level is more than 4 meter below soil surface, the conditions were so dry that there was mostly not available water to assess element leaching (there are only data for NO<sub>3</sub>, NH<sub>4</sub> and Cl). Consequently, we could not derive a budget for most of the elements at this plot.

At Hardenberg and specifically Zeist, the leaching of NO<sub>3</sub> is larger than the input, indicating the occurrence of nitrification of the incoming  $NH_4$  to  $NO_3$  and subsequent leaching. Comparing the total N input and output shows an average leaching rate of 50% for Hardenberg (N leaching is 1629 mol, ha-1.yr-1 and N input is 3214 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>) and 67% for Zeist (N leaching is 1486 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> and N input is 2210 mol. ha<sup>-1</sup>.yr<sup>-1</sup>). Considering the fact that Cl and Na generally behave as a tracer, however, this quantification seems correct for Hardenberg, where the input and output of Cl and Na is comparable, but an underestimate for Zeist (net retention of Cl). This would imply that at Zeist the system is almost complete nitrogen saturated. Inversely, the results for Dwingeloo seem to show an almost complete N retention (N leaching is -9 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup> and N input is 3118 mol<sub>c</sub>.ha<sup>-1</sup>.yr<sup>-1</sup>). However, the output of Cl and Na is also about 25% of the input, implying that this result may be biased. The calculation of the hydrological fluxes was difficult due to the large differences in height on the site caused by former sand dunes. Nevertheless, it is clear that Dwingeloo shows a different behaviour regarding the fate of N, which is most likely due to a thick layer of heath and mosses, acting as a sink for N. In summary, it can be concluded that there is likely to be a nearly complete N retention at Dwingeloo, an intermediate N retention (near 50%) at Hardenberg and hardly any N retention at Zeist.

The S budgets at Hardenberg and Dwingeloo seem to indicate retention of S, but the results are ambiguous for Dwingeloo, considering the fate of Cl and Na at this plot. This also holds for the behaviour of all base cations in this plot. At Hardenberg, there seems to be retention of base cations. On the other hand, there is a strong mobilization of Al in the soil profile at this plot (input form the atmosphere is

negligible), with Al leaching being nearly equal to the leaching of  $SO_4$  and  $NO_3$ . This illustrates that Al release, which is the clearest signal of soil acidification, is mainly due to the external input of  $SO_4$  and  $NO_3$ . At Dwingeloo, there is hardly any soil acidification due to the retention of S and N in the soil profile.

Element	Type of flux	Flux (mol <sub>c</sub> .ha <sup>-1</sup> .yr	1)	
		Hardenberg	Zeist	Dwingeloo
		Douglas-fir	Oak	Scots pine
SO <sub>4</sub>	Total deposition	1170	1034	942
	Throughfall	1141	910	902
	Leaching at 80 cm	837	_1	247
	Soil budget	304	-	655
	Total Budget	333	-	287
NO <sub>3</sub>	Total deposition	1005	456	838
	Throughfall	981	402	803
	Leaching at 80 cm	1624	1342	-63
	Soil budget	-643	-941	869
	Total Budget	-918	-886	-31
NH4	Total deposition	2209	1754	2280
	Throughfall	2146	1191	1835
	Leaching at 80 cm	5	144	54
	Soil budget	2141	1047	1780
	Total Budget	2204	1610	500
Al	Leaching at 80 cm	2608	_1	209
Ca	Total deposition	608	181	327
	Throughfall	322	385	282
	Leaching at 80 cm	184	_1	103
	Soil budget	139	-	177
	Total Budget	424	-	150
Mg	Total deposition	548	303	473
0	Throughfall	423	457	417
	Leaching at 80 cm	359	_1	77
	Soil budget	64	-	339
	Total Budget	189	-	135
Κ	Total deposition	404	75	161
	Throughfall	332	943	569
	Leaching at 80 cm	30	_1	24
	Soil budget	302	-	544
	Total Budget	374	-	-383
Na	Total deposition	1501	1355	2059
	Throughfall	1465	1192	1974
	Leaching at 80 cm	1495	_1	551
	Soil budget	-29	-	1423
	Total Budget	7	-	635
Cl	Total deposition	1738	1748	2493
	Throughfall	1696	1538	2391
	Leaching at 80 cm	1701	893	553
	Soil budget	-5	645	1836
	Total Budget	37	855	657

Table 6.7 Average yearly nutrient fluxes and nutrient budgets for the period 2003-2005 for Hardenberg, Zeist and Dwingeloo

<sup>1</sup> In case of a – there was not enough soil solution to measure the concentrations and derive a flux for the considered element. The reliability of the other elements can be questioned at this site.

## 7 Conclusions

More than 10 years of forest monitoring have shown the following trends

1. A significant worsening of the crown condition of Douglas fir and Oak, in terms of an increase in defoliation and a decrease in vitality class, over the period 1990 - 2006. For Scots pine, there is a recovery at all level I plots, but at level 2 plots there has been a decline around 1993 and there are no signs of recovery until 2006.

2. A significant decrease in  $SO_4$  deposition at all 4 plots (Hardenberg, Zeist, Dwingeloo and Speuld), whereas downward trends in nitrogen compounds are only significant for Zeist and Dwingeloo for the period 1995-2005. There is significant increase in N/S ratio on all plots but no significant trend in the NH<sub>4</sub>/NO<sub>3</sub> ratio.

4. A significant decrease in the N-content and S-content in the needles of Douglas-fir and Pine over the period 1990-2005. For Oak, the changes are not significant. The Mg-content increases significantly for all tree species, the K-content in Douglas-fir only, whereas changes in P- and Ca-content are not significant.

5. A significant decrease in  $SO_4$  concentrations in soil solution in both the topsoil and subsoil for all tree species over the period 1990-2001. Furthermore, there is a highly significant decrease in Al and Mg concentrations in the subsoil for all tree species, except for Al under Oak. The NO<sub>3</sub> concentration decreases only significantly under Pine, specifically in the subsoil.

Other conclusions are:

1 The changes in stem wood volume are generally much higher for Douglas fir (near 25 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup>) than for Scots pine and Oak (8-10 m<sup>3</sup>.ha<sup>-1</sup>.yr<sup>-1</sup>), with related carbon pool changes near 6 ton C.ha<sup>-1</sup>.yr<sup>-1</sup> for Douglas fir and near 2.5 ton C.ha<sup>-1</sup>.yr<sup>-1</sup> for both Pine and Oak.

2 The species composition in 2000 and 2005 shows that the (i) plots are poor in species, (ii) differences in species composition can largely be explained from the difference in dominant tree species (Scots pine, Douglas-fir or Oak), (iii) species composition indicates acid and nutrient poor conditions, (iv) changes between 2000 and 2005 are negligible, implying no evident abiotic changes.

3 There is likely to be nearly complete N retention at Dwingeloo, an intermediate N retention (near 50%) at Hardenberg and hardly any N retention at Zeist. At Hardenberg, there seems to be retention of sulphur and base cations and a strong mobilization, indicating soil acidification due to the external input of  $SO_4$  and  $NO_3$ . At Dwingeloo, there is hardly any soil acidification due to the retention of S and N in the soil profile.

There are strong correlations between deposition and the chemical composition of foliage, specifically between the deposition and foliar content of S, Mg and K for Oak and Pine. Correlations between nutrient deposition and soil solution chemistry appear to be much less. The results indicate a decrease in acidification, related to a decrease in  $SO_4$  and Al concentration, but the problems with N deposition remain, specifically unbalanced nutrient ratios in the foliage, due to the ongoing high N deposition, even though the trend is downward.

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Year	С	Ν	Р	S	Κ	Mg	Ca	Na	Mn	Al	Fe	Cu	Zn	В
	mg/g							µg/g						
Level 2														
1990		21	1.6		6.5	0.7	2.6							
1992	372	19	1.4	1.2	6.2	0.7	1.9	125		160		4.0	56	
1993		19	1.5	1.7	6.6	0.7	1.9	150		205		4.6	56	
1994		21	1.6	1.4	6.7	0.6	2.0	125		205		4.8	56	
1995		17	1.2	1.2	6.1	0.7	1.6	140	214	164	55	3.4	48	
1996		19	1.3		5.9	0.7	1.5	120	322	169		3.8	51	
1997		21	1.4	1.5	6.3	0.7	1.8	200		223	87	4.8	122	
1999		19	1.3	1.2	6.6	0.8	1.9		252	165	61	4.3	50	
2001		20	1.6	1.2	6.5	0.9	2.0		243	175	66	5.3	52	24
2003		17	1.2	1.0	5.9	0.8	1.7	48	202	168	51	3.7	45	
2005		17	1.4	1.0	6.9	0.8	2.2	52	255	174	44	2.2	57	
national														
1990		21	1.4		6.0	0.7	2.4					4.3		
1995		19	1.3	1.2	6.1	0.7	1.8	162	234	204	66	3.7	47	
2000		16	0.9	0.9	4.8	0.6	2.2	184	196	207	78	4.8	39	

Appendix 2 Average foliar contents per tree species over de period 1990 - 2003

Douglas	-fir

Year	С	Ν	Р	S	Κ	Mg	Ca	Na	Mn	Al	Fe	Cu	Zn	В
	mg/g							μg/g						
Level 2														
1990		23	1.4		6.2	1.5	3.0					3.4		
1992	397	19	1.3	1.5	6.7	1.3	2.3	125		192		3.8	20	
1993		20	1.3	1.8	5.6	1.3	3.0	150		236		4.5	21	
1994		19	1.4	1.5	7.0	1.4	2.6	100		195		4.9	22	
1995		18	1.2	1.4	6.7	1.4	2.4	120	487	245	72	3.9	24	
1996		19	1.4		7.6	1.4	2.2	100	83	180		5.2	31	
1997		19	1.3	1.5	6.7	1.4	2.5	150		248	88	4.2	79	
1999		18	1.3	1.5	7.8	1.8	2.8	125	435	189	77	4.5	27	
2001		19	1.2	1.2	6.6	1.6	2.8	150	366	148	56	4.5	24	36
2003		18	1.1	1.2	7.4	1.4	1.9	27	379	188	60	3.2	27	
2005		17	1.2	1.1	8.0	1.6	2.7	42	555	159	65	4.3	35	
national														
1990		22	1.2		6.2	1.5	4.1					3.6		
1995		19	1.2	1.4	6.5	1.4	2.6	137	375	256	88	4.0	26	
2000		20	0.9	1.2	4.7	1.1	4.4	282	352	244	121	6.1	26	

Oak
-----

V	C	NT	D	C	IZ.	м	C	NT	м	A 1	P	C	7	р
Year	С	Ν	Р	S	Κ	Mg	Ca	Na	Mn	Al	Fe	Cu	Zn	В
	mg/g							µg/g						
Level 2														
1990		28	1.8		8.8	1.4	3.9							
1992	434	26	1.8	1.6	7.1	1.4	3.6	150		73		7.0	29	
1993		25	2.0	1.6	7.1	1.2	4.1	100		98		7.3	28	
1994		24	1.8	1.6	8.9	1.5	3.7	133		60		8.3	22	
1995		25	1.6	1.7	7.6	1.7	3.9	125	688	80	96	7.3	21	
1996		26	1.9		8.9	1.5	2.5	133		50		7.7	20	
1997		29	1.9	1.9	9.9	1.4	3.1	100		65	110	8.3	25	
1999		29	1.7	1.4	8.9	1.6	3.0	150	626	65	115	8.2	22	
2001		28	2.1	1.7	9.6	1.8	4.3		655	45	111	9.3	28	38
2005		24	2.4	1.3	11	1.8	4.1	171	321	74	119	4.3	27	
national														
1990		28	1.6		9.2	1.5	4.9							
1995		25	1.5	1.7	8.9	1.7	5.2	139	621	79	126	7.6	25	
2000		52	1.6	1.5	8.3	1.5	5.7	169	740	85	153	8.9	22	

# Appendix 3 Average element concentrations in the soil solution per tree species over de period 1990 – 2001

topsoi			<u>c:</u>	Δ 1	E	C	м	V	NT	NITT	м	NO	$C^{1}$	50	DO
Year	Species	pH 27	Si	Al	Fe	Ca	Mg	K	Na	NH4	Mn	$NO_3$	Cl	SO <sub>4</sub>	$PO_4$
1990	Pine	3.7	1.39	0.68	0.02	0.36	0.18	0.20	0.74	0.18	0.01	0.39	0.99	0.95	0.006
	Douglas	3.8	1.65	1.63	0.04	0.57	0.49	0.32	2.04	0.42	0.03	0.66	2.71	1.87	0.005
	Oak	3.7 3.7	0.96	0.44	0.01	0.30	0.21	0.41	0.66	0.17	0.03	0.49	1.03	0.64	0.005
1002	All		1.33	0.92	0.02	0.41	0.29	0.31	1.14	0.26	0.02	0.51	1.58	1.15	0.006
1992	Pine	3.8 3.5	1.41 1.46	0.65	0.05	0.54 0.52	0.20 0.27	0.17	0.56 1.03	0.26 0.48	0.01	0.50	0.58	1.02 1.39	0.005 0.014
	Douglas Oak	3.5 3.8	0.73	0.99 0.24	0.03 0.03	0.32	0.27	0.13 0.25	0.29	0.46	0.02 0.03	$0.68 \\ 0.74$	1.23 0.30	0.42	0.014
	All	3.8 3.7	1.20	0.24	0.03	0.31	0.15	0.25	0.29	0.10	0.03	0.74	0.30	0.42	0.021
1993	Pine	3.7 3.9	1.20	0.65	0.04	0.40	0.21		0.63	0.30	0.02	0.84	0.71	0.94	0.015
1995		3.9 3.5	1.29	0.47	0.03	0.41	0.22	0.13 0.22	0.62	0.18	0.01		0.80 1.34	1.11	0.015
	Douglas Oalr	3.5 3.8	0.74		0.02	0.48		0.22	0.75	0.29	0.01	0.46	0.57	0.46	0.007
	Oak All	3.8 3.7	1.13	0.35 0.54	0.02	0.32	0.21 0.24	0.22	0.44	0.15	0.04	0.66 0.49	0.97	0.40	0.022
1994	Pine	4.3	0.86	0.34	0.02	0.41	0.24	0.19	0.00	0.21	0.02	0.49	0.92	0.80	0.013
1994		4.3 3.7	0.80	0.20	0.07	0.34	0.13	0.15	0.20	0.24	0.01	0.41	0.31	0.70	0.001
	Douglas Oak	4.2	0.70	0.43	0.04	0.38	0.18	0.13	0.18	0.30	0.01	0.43	0.49	0.73	0.004
	All	4.1	0.41	0.24	0.05	0.19	0.07	0.14	0.00	0.11	0.01	0.25	0.14	0.55	0.005
1995	Pine	4.1 3.9	0.88	0.32	0.03	0.30	0.13	0.14	0.13	0.24	0.01	0.30	0.31	0.38	0.003
1775	Douglas	3.7	1.14	0.32	0.03	0.22	0.14	0.07	0.42	0.11	0.00	0.2)	0.42	1.71	0.001
	Oak	3.8	0.70	0.47	0.03	0.27	0.22	0.19	0.30	0.08	0.02	0.63	0.38	0.59	0.001
	All	3.8	0.92	0.26	0.02	0.27	0.18	0.19	0.50	0.10	0.03	0.03	0.45	1.10	0.007
1996	Pine	4.2	1.63	0.79	0.03	0.40	0.10	0.18	0.20	0.30	0.02	1.03	0.45	0.62	0.003
1770	Douglas	3.6	1.76	1.18	0.03	0.51	0.42	0.15	0.20	0.26	0.01	0.86	1.28	1.10	0.004
	Oak	3.8	1.29	0.46	0.02	0.30	0.12	0.25	0.12	0.17	0.02	0.87	0.35	0.46	0.023
	All	3.9	1.56	0.81	0.02	0.40	0.31	0.19	0.22	0.25	0.02	0.92	0.71	0.73	0.012
1997	Pine	4.0	1.56	0.42	0.04	0.36	0.19	0.15	0.74	0.27	0.01	0.37	0.95	0.77	0.005
1777	Douglas	3.6	1.67	0.88	0.05	0.56	0.32	0.15	1.11	0.28	0.03	0.83	1.11	1.22	0.01
	Oak	3.6	1.12	0.46	0.03	0.40	0.24	0.36	0.47	0.18	0.03	1.01	0.53	0.60	0.024
	All	3.7	1.47	0.60	0.04	0.44	0.25	0.21	0.80	0.25	0.02	0.72	0.89	0.88	0.012
1998	Pine	5.1	0.11	0.19	0.03	0.50	0.12	0.09	0.48	0.07	0.00	0.34	0.43	0.52	0
	Douglas	4.1	0.15	0.69	0.03	0.61	0.26	0.11	1.54	0.09	0.02	0.63	1.21	1.10	0.001
	Oak	4.3	0.10	0.30	0.03	0.58	0.20	0.24	0.33	0.11	0.02	0.84	0.44	0.43	0.01
	All	4.5	0.12	0.40	0.03	0.56	0.19	0.14	0.82	0.09	0.01	0.59	0.71	0.70	0.004
1999	Pine	4.7	0.90	0.21	0.07	0.28	0.09	0.08	0.34	0.04	0.00	0.15	0.43	0.13	0.002
	Douglas	4.0	1.24	0.46	0.04	0.62	0.23	0.10	0.48	0.08	0.02	0.48	0.63	0.22	0.003
	Oak	4.6	0.86	0.24	0.04	0.36	0.12	0.16	0.28	0.04	0.01	0.35	0.37	0.11	0.01
	All	4.4	1.01	0.31	0.05	0.42	0.15	0.11	0.37	0.06	0.01	0.32	0.49	0.16	0.005
2000	Pine	4.8	*	0.27	0.04	0.33	0.10	0.09	0.52	0.03	0.01	0.16	0.50	0.14	0
	Douglas	3.6	*	0.79	0.04	0.65	0.30	0.08	1.03	0.16	0.03	1.13	1.12	0.26	0
	Oak	4.1	*	0.28	0.03	0.49	0.18	0.18	0.34	0.07	0.02	0.56	0.43	0.11	0.011
	All	4.2	*	0.46	0.04	0.49	0.20	0.11	0.65	0.09	0.02	0.62	0.71	0.17	0.003
2001	Pine	4.0	*	0.33	0.04	0.10	0.07	0.06	0.46	0.02	0.00	0.17	0.38	0.10	0.002
	Douglas	3.4	*	1.92	0.03	0.44	0.45	0.10	1.34	0.20	0.07	1.53	1.61	0.30	0
	Oak	4.0	*	0.35	0.02	0.13	0.13	0.11	0.36	0.04	0.02	0.34	0.34	0.09	0.003
	All	3.8	*	0.79	0.03	0.21	0.20	0.08	0.68	0.08	0.03	0.60	0.72	0.16	0.002
All	Pine	4.3	1.09	0.41	0.04	0.36	0.15	0.12	0.48	0.16	0.01	0.37	0.58	0.60	0.004
	Douglas	3.7	1.22	0.86	0.03	0.52	0.30	0.14	0.95	0.25	0.02	0.71	1.18	1.02	0.005
	Oak	4.0	0.77	0.33	0.03	0.34	0.18	0.23	0.33	0.12	0.03	0.62	0.45	0.40	0.014
	All	4.0	1.04	0.54	0.03	0.41	0.21	0.16	0.60	0.17	0.02	0.56	0.74	0.68	0.007

subsoi		T T	<u> </u>	A 1	Г	6		17	NT	N 17 7		NO	<u>C1</u>		DC
Year	Species	pH 2.0	Si	Al	Fe	Ca	Mg	K	Na	NH <sub>4</sub>	Mn	NO <sub>3</sub>	Cl	SO <sub>4</sub>	PO <sub>4</sub>
1990	Pine	3.9	1.38	0.83	0.01	0.37	0.21	0.12	0.64	0.08	0.01	0.61	0.92	0.88	0.001
	Douglas	3.8	1.24	1.04	0.11	0.25	0.26	0.09	1.41	0.08	0.07	0.65	1.58	1.18	0
	Oak	4.0	1.13	0.29	0.03	0.27	0.22	0.20	0.67	0.11	0.08	0.35	0.81	0.79	0.004
1000	All	3.9	1.25	0.72	0.05	0.29	0.23	0.13	0.91	0.09	0.05	0.54	1.11	0.95	0.001
1992	Pine	4.1	1.36	0.79	0.00	0.31	0.16	0.06	0.67	0.05	0.02	0.56	0.74	0.86	0
	Douglas	4.0	1.60	1.72	0.01	0.53	0.31	0.08	1.56	0.11	0.04	0.97	1.64	1.63	0.001
	Oak	4.2	1.04	0.29	0.00	0.30	0.19	0.09	0.38	0.10	0.06	0.70	0.33	0.43	0
1000	All	4.1	1.33	0.94	0.01	0.38	0.22	0.08	0.87	0.09	0.04	0.74	0.90	0.97	0
1993	Pine	4.2	1.26	0.44	0.01	0.34	0.15	0.08	0.51	0.07	0.02	0.25	0.50	0.86	0.001
	Douglas	4.0	1.56	1.36	0.01	0.36	0.25	0.21	1.02	0.09	0.02	0.58	1.21	1.43	0.002
	Oak	4.2	0.84	0.30	0.01	0.14	0.14	0.17	0.16	0.08	0.06	0.26	0.32	0.43	0.003
1004	All	4.1	1.22	0.70	0.01	0.28	0.18	0.15	0.56	0.08	0.03	0.37	0.67	0.91	0.002
1994	Pine	4.5	0.93	0.32	0.01	0.34	0.13	0.10	0.17	0.08	0.01	0.31	0.26	0.69	0.003
	Douglas	4.0	1.58	0.96	0.03	0.53	0.26	0.04	0.32	0.07	0.03	0.51	0.46	1.45	0.002
	Oak	4.5	0.65	0.24	0.01	0.28	0.10	0.06	0.07	0.07	0.04	0.18	0.14	0.37	0.003
1005	All	4.3	1.05	0.51	0.01	0.38	0.17	0.07	0.19	0.07	0.03	0.33	0.29	0.84	0.003
1995	Pine	4.2	0.92	0.30	0.01	0.17	0.12	0.04	0.30	0.04	0.01	0.28	0.20	0.83	0
	Douglas	4.1	1.33	0.76	0.07	0.26	0.19	0.04	0.39	0.06	0.02	0.40	0.78	1.09	0
	Oak	4.2	1.07	0.29	0.04	0.23	0.13	0.02	0.28	0.02	0.03	0.24	0.13	0.71	0.003
1007	All D'	4.1	1.11	0.46	0.04	0.22	0.15	0.03	0.33	0.04	0.02	0.31	0.32	0.89	0.001
1996	Pine	4.3	1.25	0.75	0.01	0.32	0.16	0.08	0.17	0.08	0.02	0.77	0.37	0.52	0.003
	Douglas	4.1	1.56	0.99	0.02	0.36	0.26	0.07	0.26	0.13	0.03	0.77	0.81	0.73	0.003
	Oak	4.2	1.45	0.54	0.01	0.31	0.25	0.10	0.17	0.06	0.07	0.62	0.51	0.42	0.008
1007	All D'	4.2	1.42	0.76	0.01	0.33	0.23	0.09	0.20	0.09	0.04	0.72	0.56	0.56	0.005
1997	Pine	4.5	1.14	0.22	0.03	0.38	0.15	0.07	0.74	0.16	0.01	0.34	0.54	0.74	0.002
	Douglas	4.3	1.62	0.71	0.16	0.73	0.30	0.09	1.13	0.18	0.08	0.84	1.74	1.17	0.004
	Oak	4.3	1.43	0.31	0.07	0.41	0.19	0.12	0.58	0.15	0.04	0.53	0.47	0.71	0.004
1000	All D'	4.4	1.39	0.42	0.09	0.51	0.22	0.09	0.83	0.16	0.04	0.57	0.95	0.89	0.004
1998	Pine	5.1	0.11	0.11	0.01	0.43	0.12	0.05	0.51	0.03	0.01	0.30	0.44	0.63	0
	Douglas	4.5	0.14	0.56	0.02	0.61	0.23	0.08	1.28	0.05	0.04	0.66	1.29	1.06	0
	Oak	4.7	0.13	0.22	0.01	0.42	0.17	0.10	0.44	0.05	0.03	0.55	0.48	0.52	0.002
1000	All	4.8	0.13	0.30	0.01	0.49	0.17	0.08	0.77	0.04	0.02	0.50	0.75	0.75	0
1999	Pine	4.8	0.93	0.13	0.04	0.29	0.10	0.09	0.32	0.02	0.01	0.16	0.48	0.16	0.001
	Douglas	4.4 5.4	1.44	0.37	0.07	0.60	0.19	0.06	0.65	0.05	0.03	0.30	0.84	0.27	0.001
	Oak	5.4	1.09	0.27	0.14	0.37	0.12	0.07	0.30	0.04	0.02	0.17	0.40	0.13	0.002
2000	All D'	4.8	1.16 *	0.26	0.08	0.42	0.14	0.07	0.43	0.04	0.02	0.21	0.59	0.19	0.001
2000	Pine	4.2	*	0.24	0.01	0.27	0.11	0.04	0.59	0.04	0.01	0.25	0.62	0.16	0
	Douglas	4.2		0.68	0.04	0.63	0.24	0.06	1.31	0.12	0.04	0.73	1.15	0.33	0
	Oak	4.7	*	0.21	0.04	0.29	0.12	0.07	0.37	0.09	0.03	0.25	0.35	0.16	0
2001	All D'	4.3	*	0.39	0.03	0.41	0.16	0.05	0.78	0.09	0.02	0.42	0.70	0.22	0
2001	Pine	4.2	*	0.31	0.01	0.07	0.07	0.05	0.44	0.01	0.01	0.10	0.38	0.15	0
	Douglas	4.0	*	1.18	0.01	0.26	0.29	0.08	0.91	0.03	0.12	0.59	0.77	0.23	0
	Oak	4.4	*	0.16	0.00	0.10	0.11	0.08	0.42	0.03	0.03	0.10	0.38	0.12	0
A 11	All D'	4.2	*	0.52	0.01	0.13	0.15	0.07	0.57	0.02	0.05	0.24	0.49	0.16	0
All	Pine	4.4	1.01	0.39	0.01	0.31	0.14	0.07	0.47	0.06	0.01	0.36	0.51	0.58	0.001
	Douglas	4.2	1.32	0.90	0.05	0.49	0.25	0.08	0.95	0.09	0.04	0.64	1.16	0.97	0.001
	Oak	4.4	0.98	0.29	0.03	0.30	0.16	0.10	0.35	0.08	0.04	0.38	0.40	0.44	0.003
	All	4.3	1.11	0.53	0.03	0.37	0.18	0.08	0.59	0.08	0.03	0.46	0.69	0.67	0.002

## Appendix 4 Estimating plot mean average soil solution chemistry data from composite samples in the presence of non-response

#### Non-response

The response variable is the concentration of a composite sample. The predictors are the proportions of the composite sample originating from the strata, symbolized by  $x_j = 1...L$ , where L is the number of strata or predictors. The predictor  $x_j$  has value 0 if the composite sample contains no soil solution from stratum *j*, else value 1/m, where *m* is the total number of strata from which soil solution was collected to form the composite (number of aliquots of composite). To estimate the plot-mean concentration, the following relation is assumed:

$$\mathbf{y}_{i} = \sum_{j=1}^{L} \boldsymbol{\beta}_{j} \mathbf{x}_{j} + \boldsymbol{\varepsilon}_{i}, \qquad (A4.1)$$

where  $\beta_j$  is the regression coefficient (mean concentration) of stratum *j* to be estimated, and  $\varepsilon_i$  is the residual. Moreover, it is assumed that the  $\varepsilon$ 's are independent, and have normal distribution with expectation 0 and variance  $\sigma^2$ . In matrix-notation Eq. (A4.1) becomes

$$\mathbf{y} = \mathbf{X}\boldsymbol{\beta} + \boldsymbol{\varepsilon} \tag{A4.2}$$

where **y** is the *n*-vector with concentrations of the composites, **X** is the  $(n \ge L)$ -matrix with values for the predictors (design-matrix),  $\beta$  is the *L*-vector with regression coefficients, and  $\varepsilon$  is the *n*-vector with residuals.

The regression coefficients are estimated by (Draper & Smith, 1981):

$$\mathbf{b} = (\mathbf{X}^{\mathrm{T}}\mathbf{X})^{-1}\mathbf{X}^{\mathrm{T}}\mathbf{y}, \tag{A4.3}$$

The mean concentration for the plot is estimated by the expectation of *y* when all  $x_j$ 's are equal to 1/L:

$$\hat{\mathbf{y}}_0 = \mathbf{E}[\mathbf{y}] = \mathbf{x}_0^{\mathrm{T}} \mathbf{b}, \tag{A4.4}$$

where  $\mathbf{x}_0$  is the *L*-vector with values 1/L for all entries.

The variance of  $\hat{y}_0$  is estimated by

$$\mathbf{v}(\hat{\mathbf{y}}_0) = \boldsymbol{\sigma}^2 \mathbf{x}_0^{\mathrm{T}} (\mathbf{X}^{\mathrm{T}} \mathbf{X})^{-1} \mathbf{x}_0.$$
(A4.5)

Note that this regression-based estimator requires the number of composites  $\geq$  number of strata. If the number of composites equals the number of strata, the variance (Eq. A4.5) can not be estimated.

## Aliased predictors

The method described above fails when a predictor is a linear combination of one or more of the other predictors. The aliased predictor can not be included in the model, there is no additional information in this predictor. In statistical terms this is referred to as aliasing. Two situations must be distinguished:

- aliased predictor that is identical to another predictor
- aliased predictor that is not identical to another predictor

*Identical aliased predictors.* In the first case two (or even more) columns in the designmatrix  $\mathbf{X}$  are identical. In this case we can not obtain estimates for each stratum separately, but we can obtain estimates for groups of strata. This can be done by reducing the design-matrix  $\mathbf{X}$ : identical columns are substituted by one column with entries equal to the sum of the entries in the original columns. In the example shown hereafter the first and third column in the 4 x 3 design-matrix  $\mathbf{X}$  are identical. The entries of these columns are added, which results in the first column of  $\mathbf{X}_r$ .

X		$\mathbf{X}_{r}$
$\begin{bmatrix} \frac{1}{3} & \frac{1}{3} & \frac{1}{3} \end{bmatrix}$	] [	2/3 1/3
1/3 1/3 1/3		$\frac{2}{3}\frac{1}{3}$
$\frac{1}{2}0\frac{1}{2}$		1 0
$\begin{bmatrix} \frac{1}{3} \frac{1}{3} \frac{1}{3} \frac{1}{3} \\ \frac{1}{3} \frac{1}{3} \frac{1}{3} \frac{1}{3} \\ \frac{1}{2} 0 \frac{1}{2} \\ \frac{1}{3} \frac{1}{3} \frac{1}{3} \frac{1}{3} \end{bmatrix}$		<sup>2</sup> / <sub>3</sub> <sup>1</sup> / <sub>3</sub>

In this example the first regression coefficient is an estimate of the mean of stratum 1 + stratum 3. To estimate the plot mean concentration and its variance we must also reduce the vector (3 x 1)-vector  $\mathbf{x}_0$  to a (2 x 1)-vector  $\mathbf{x}_{0r}$ 

$$\begin{array}{c} \mathbf{x}_{0} & \mathbf{x}_{0,\mathbf{r}} \\ \begin{bmatrix} y_{3} \\ y_{3} \\ y_{3} \end{bmatrix} \rightarrow \begin{bmatrix} 2y_{3} \\ y_{3} \end{bmatrix} \\ \end{array}$$
(A4.7)

The plot mean concentration and its variance can then be estimated by substituting  $\mathbf{X}_{r}$  and  $\mathbf{x}_{0,r}$  in Eqs. (A4.3) – (A4.5).

Note that in case of no non-response design-matrix **X** reduces to a  $(n \ge 1)$ -vector, and the regression-estimator (Eq. A4.5) equals the unweighted mean of the composite concentrations (Eq. 2.1). Also, the variance estimators Eq. (A4.7) and Eq. (2.2) are equivalent.

Aliased predictors, not identical An example of the second situation, i.e. an aliased predictor that is not identical to another predictor, is the following design-matrix:

 $\begin{bmatrix} 0 & 0 & \frac{1}{5} & \frac{1}{5} & 0 \\ \frac{1}{4} & \frac{1}{4} & 0 & \frac{1}{4} & \frac{1}{4} \\ \frac{1}{4} & \frac{1}{4} & 0 & \frac{1}{4} & \frac{1}{4} \\ 0 & \frac{1}{3} & 0 & \frac{1}{3} & \frac{1}{3} \\ \frac{1}{3} & 0 & \frac{1}{3} & \frac{1}{3} & 0 \\ 0 & 1 & 0 & 0 & 0 \end{bmatrix}$ 

In this example x[5] = -x[3] + x[4]. In other terms x[5] is aliased with x[3] and x[4], and can not be fitted. At first sight the problem can be solved by summing x[3] and x[4], or summing x[4] and x[5]). However, the problem is that the two predictors being summed are not identical, i.e. the corresponding strata are not equally represented in the composites. In this case, there is no solution to the problem of aliased predictors. In this case the plot-mean is estimated by the unweighted mean of the composite concentrations (Eq. 2.1). We do not know the accuracy (bias + variance) of this unweighted mean.

#### Number of composites smaller than number of strata

The regression-based estimator fails when the number of composites is smaller than the number of strata. In this case the plot-mean is estimated by the unweighted mean of the composite concentrations, with a missing value for its variance.

If one or more strata are missing in all composites, then the mean for the sampled part of the plot is estimated by the regression-based estimator, or if this fails (aliased predictors that are not identical, or number of composites smaller than number of strata) by the unweighted mean.

(A4.8)