Tree mineral nutrition is deteriorating in Europe

MA T H I E U J ON AR D 1 , A L F R E D F U¨ RS T 2 , A RNE V ERSTRAETEN 3 , A NNE THIMONI E R 4 , VOLKMAR T IMMERMANN 5 , N ENA D POTO Cˇ I C' 6 , P ET ER W A LDNER 4 , S UE BENHAM 7 ,

KAR I N H ANSE N 8 , P A¨ IVI M ERIL A¨ 9 , QUENTIN PONETTE 1 , A N A C D E L A C R U Z 1 0 , P ET ER ROS K AMS 3 , M ANUEL N ICOLAS 11 , L UC CR OIS E´ 11 , M ORTE N I NG ER SLE V 1 2 , G I O R G I O

MATT EUC C I 1 3 , B RUNO D E CINTI 1 4 , M ARC O BAS C IE TT O 1 4 and PASI RAUTIO 9

1*UCL-ELI, Universite´ catholique de Louvain, Earth and Life Institute, Croix du Sud 2, L7.05.09, Louvain-la-Neuve BE-1348, Belgium,* 2*Federal Research Centre for Forests, BFW, A-1131 Vienna, Austria,* 3*Research Institute for Nature and Forest, Gaverstraat 35, Geraardsbergen B-9500, Belgium,* 4*WSL, Swiss Federal Institute for Forest, Snow and Landscape Research, Birmensdorf CH-8903, Switzerland,* 5*Norwegian Forest and Landscape Institute, Pb 115, NO-1431 Aas, Norway,* 6*Department of Ecology, Croatian Forest Research Institute, Cvjetno naselje 41, HR-10450 Jastrebarsko, Croatia,* 7*Forest Research, Alice Holt*

*Lodge, Wrecclesham, Farnham, Surrey GU10 4LH, United Kingdom,* 8*IVL Swedish Environmental Research Institute, Stockholm SE-100 31, Sweden,* 9*METLA, Finnish Forest Research Institute, PL 18, Vantaa FI-01301, Finland,* 10*INIA, Centro de Investigaci*'*on Forestal, Ctra. de la Corun*~*a km. 7.5, Madrid ES-28040, Spain,* 11*De´partement Recherche et De´veloppement, ONF, Office National des For*^*ets, B*^*atiment B, Boulevard de Constance, Fontainebleau F-77300, France,* 12*Department of Geosciences and Natural Resource Management, University of Copenhagen, Rolighedsvej 23, Frederiksberg DK-1958, Denmark,* 13*Istituto per i Sistemi Agricoli e Forestali del Mediterraneo - U.O.S. Rende, Via Cavour 4-6, Rende I-87030, Italy,* 14*CNR, Istituto di Biologia*

*Agroambientale e Forestale - U.O.S. Montelibretti, Via Salaria km 29.300, Monterotondo Scalo I-00016, Italy*

# Abstract

The response of forest ecosystems to increased atmospheric CO2 is constrained by nutrient availability. It is thus crucial to account for nutrient limitation when studying the forest response to climate change. The objectives of this study were to describe the nutritional status of the main European tree species, to identify growth-limiting nutrients and to assess changes in tree nutrition during the past two decades. We analysed the foliar nutrition data collected during 1992–2009 on the intensive forest monitoring plots of the ICP Forests programme. Of the 22 significant tempo- ral trends that were observed in foliar nutrient concentrations, 20 were decreasing and two were increasing. Some of these trends were alarming, among which the foliar P concentration in *F. sylvatica*, *Q. Petraea* and *P. sylvestris* that sig- nificantly deteriorated during 1992–2009. In *Q. Petraea* and *P. sylvestris*, the decrease in foliar P concentration was more pronounced on plots with low foliar P status, meaning that trees with latent P deficiency could become deficient in the near future. Increased tree productivity, possibly resulting from high N deposition and from the global increase in atmospheric CO2, has led to higher nutrient demand by trees. As the soil nutrient supply was not always sufficient to meet the demands of faster growing trees, this could partly explain the deterioration of tree mineral nutrition. The results suggest that when evaluating forest carbon storage capacity and when planning to reduce CO2 emissions by increasing use of wood biomass for bioenergy, it is crucial that nutrient limitations for forest growth are considered.

*Keywords: Abies alba*, *Fagus sylvatica*, foliar nutrients, forest monitoring, *Picea abies*, *Pinus sylvestris*, *Quercus petraea*, *Quercus robur*, trend analysis

# Introduction

Global change induces simultaneous and rapid modifi- cations in many environmental factors that control dynamics of forest ecosystems (Aber *et al.*, 2001). Carbon dioxide (CO2) enrichment experiments have shown that photosynthesis is enhanced by elevated atmospheric CO2 concentration (Ceulemans *et al.*,

Correspondence: Mathieu Jonard, tel. 003210472548,

fax 003210473697, e-mail: [mathieu.jonard@uclouvain.be](mailto:mathieu.jonard@uclouvain.be)

1999). In ecosystems where nutrients and water are not limiting, this CO2 ‘fertilization effect’ results in a sub- stantial increase in net primary production (NPP), at least in the short-term (Norby *et al.*, 2005). In addition to water limitation, the response of forest ecosystems to increased atmospheric CO2 is constrained by nutrient availability, hence at the nutritionally poorest sites trees might not respond at all (Oren *et al.*, 2001). Based on a free-air CO2 enrichment experiment conducted during 11 years, Norby *et al.* (2010) showed that the enhance- ment of NPP under elevated CO2 started to decline

after 6 years due to progressive nitrogen (N) limitation. Until now, the effect of N availability has mainly been analysed because it is the limiting nutrient in many ecosystems. However, forest ecosystems located in densely populated and industrialised regions have in many cases already been enriched in N due to high atmospheric N deposition during the past decades (Aber *et al.*, 2003). In response to unbalanced N and P deposition and given the crucial role of phosphorus (P) in plant development, seminatural ecosystems are gradually moving from N to P limitation (Pen~uelas *et al.*, 2012, 2013). Other nutrients such as magnesium (Mg), potassium (K) and calcium (Ca) could also become limiting in the future or are already limiting now in nutrient-poor ecosystems (Jonard *et al.*, 2012). To avoid overestimations of the carbon (C) sequestra- tion capacity of terrestrial ecosystems, it is therefore crucial that global climate models account for the effects of nutrient limitation on the forest response to global change (Norby *et al.*, 2010; Pen~uelas *et al.*, 2013).

In Europe, the combined effects of the global enrich- ment in atmospheric CO2 and the high atmospheric N deposition have increased forest productivity during the last decades mainly because N limitation of tree growth has been progressively reduced and even disappeared in many regions (Boisvenue & Running, 2006; Solberg *et al.*, 2009; Bontemps *et al.*, 2011). Since N deposition is still high in many regions (Posch *et al.*, 2012; Reis *et al.*, 2012) and atmospheric CO2 is still ris- ing, forest productivity in Europe could continue to increase, except where water stress augments or other major nutrients become limiting. In forest ecosystems with low base saturation or P availability in the soil, the N fertilisation effect of atmospheric deposition has already resulted in nutritional imbalances (Mohren *et al.*, 1986; Katzensteiner *et al.*, 1992; Jonard *et al.*, 2010; Prietzel & Stetter, 2010).

The rise in forest productivity caused by global envi- ronmental change creates a higher nutrient demand by trees, though the increase in nutrient use efficiency may offset part of this demand (Lukac *et al.*, 2010). Given this growing nutrient demand, two scenarios are possible.

1. Trees are able to meet the enhanced nutrient demand (e.g., by improved nutrient uptake capacity or by an increased nutrient supply provided by deposition or organic matter mineralisation). We hy- pothesise that this first scenario takes place for N.
2. The higher nutrient demand cannot be met and tree nutrition progressively deteriorates. We hypothesise that this second scenario takes place for P, sulphur

(S) and base cations.

The objectives of this study were to describe the nutritional status of the main European tree species, to

identify growth-limiting nutrients and to assess changes in tree nutrition during the past two decades. We analysed the foliar nutrition data collected during 1992–2009 on the intensive forest monitoring plots of the joint EU & UNECE ICP Forests programme (Inter- national Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests working under UNECE). This dataset is unique in its scope and size, and has the further advantage of being harmon- ised among all participating countries. Factors which most probably explain the changes in foliar nutrition are discussed.

# Materials and methods

## Sampling design

Since the early nineties, the foliar nutritional status of the main tree species has been assessed regularly (usually biannually) on the intensive forest monitoring plots of the ICP Forests in Europe. In this study, we focused on the six tree species (*Fagus sylvatica* L., *Quercus petraea* (Matt.) Liebl., *Quercus robur* L., *Abies alba* Mill., *Picea abies* (L.) H. Karst, *Pinus sylvestris* L.) best represented in the ICP Forests database (tree species with more than 130 observations from at least 15 different plots). We used only data from plots for which foliar data were avail- able for at least three different samplings between 1992 and 2009. This resulted in a total of 3141 individual measurements on current-year leaves from 425 plots. The spatial distribution of the selected plots is represented per tree species in Fig. 1.

In each plot, the sample trees were selected according to the guidelines provided by the ICP Forests manual on sampling and analysis of needles and leaves (Rautio *et al.*, 2010; Rautio

& Fu¨ rst, 2013). To have a representative sample for the plot, a minimum of five trees were randomly selected among the dominant and predominant trees belonging to the most com- mon tree species. The same trees were sampled over the years, except for trees with small crown and foliage mass where alternating between two sets of sample trees avoided excess crown damage.

Foliar sampling and analysis were generally performed biannually every uneven year (Figure S1). For deciduous species, sampling was achieved during the second half of the growing season before the onset of autumnal yellowing. Coniferous species were sampled during the dormant period following the growing season of interest.

## Leaf collection

Only mature leaves or needles grown in full sun were collected from the upper third of the crown. For deciduous species, sampling was carried out on current-year leaves while both the current and 1-year-old needles were collected for coniferous species. Different sampling methods were used (climbing, tree pruner, hydraulic lift, hunting gun) depending on the tree species and the height of the sample trees but care was taken to avoid contamination (by hands or soil) and

20°0'0"W

10°0'0"W

0°0'0"

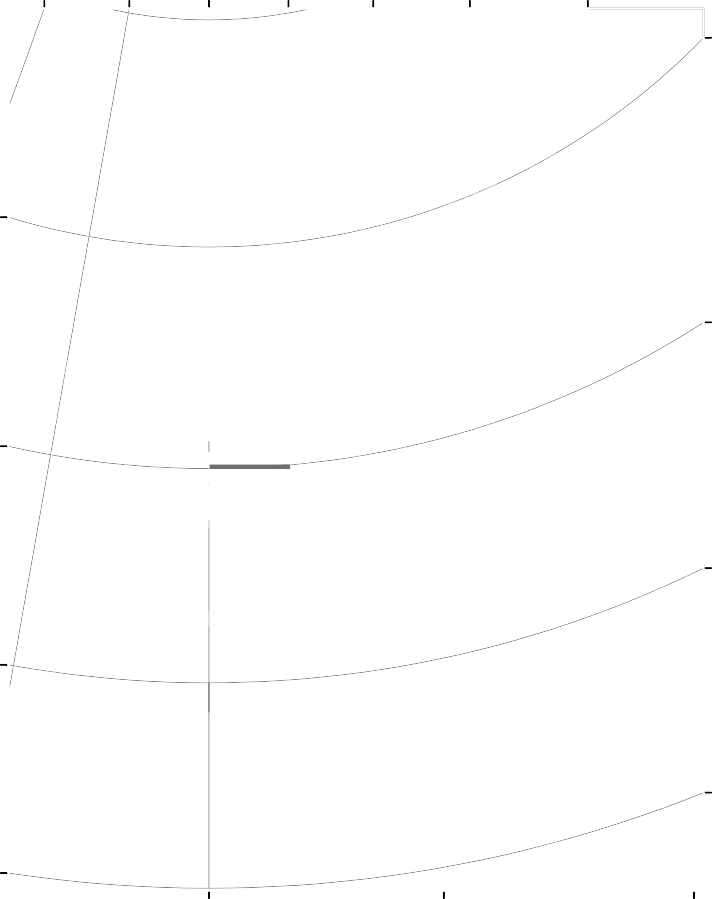
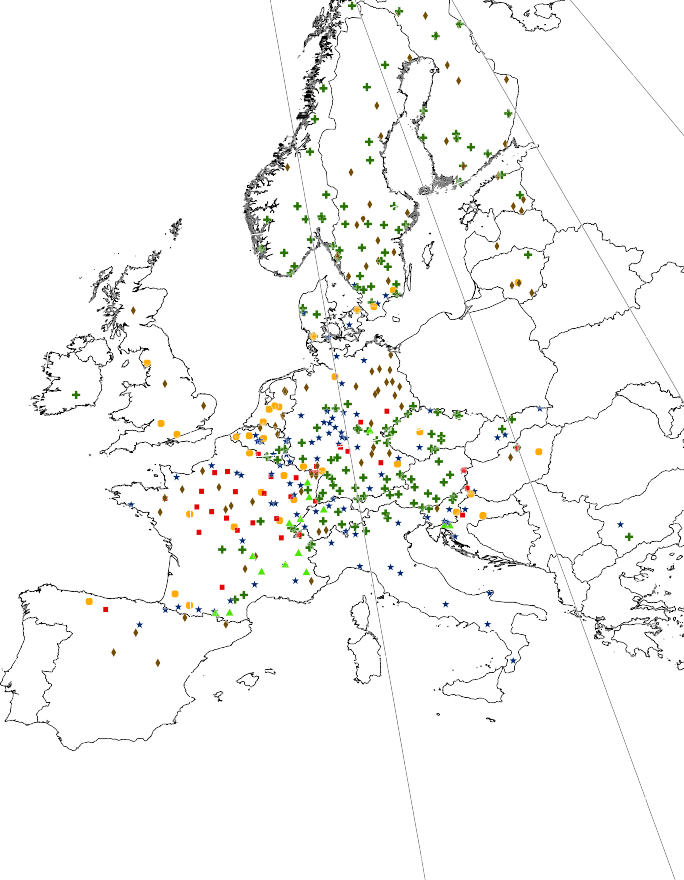
10°0'0"E

20°0'0"E

30°0'0"E

40°0'0"E

60°0'0"N



60°0'0"N

50°0'0"N

50°0'0"N

40°0'0"N

40°0'0"N

30°0'0"N

30°0'0"N

**Tree species**

0°0'0"

10°0'0"E

Kilometers

20°0'0"E

Abies alba Fagus sylvatica Picea abies Pinus sylvestris Quercus petraea Quercus robur

0 375 750 1,500

Coordinate reference system : ETRS89 lambert azimutal equal area Datum: WGS 1984

Fig. 1 Spatial distribution of the ICP forests level II intensive monitoring plots of the main tree species in Europe.

damage to the tree. The foliar samples were kept cool and dark following sampling and during transport to the laboratory.

## Pretreatment

Generally a composite sample was made by mixing equal amounts of the foliar samples from individual trees in the same plot (needle classes separately). Whether the sample was a composite sample or individual trees, the samples were divided into two subsamples: one used for the measurement of the dry mass of 100 leaves or 1000 needles and the other for chemical analyses. Both sample types were placed in paper or polyethylene bags. The samples for the determination of the

dry mass were oven-dried at 105 °C until constant weight was achieved and the samples for chemical analyses were oven- dried at a maximum temperature of 70 °C. For deciduous species, leaves were detached from twigs before drying while, for coniferous species, needles were separated from twigs after drying. Dried samples for chemical analyses were ground to obtain a fine homogeneous powder.

## Foliar analyses and quality assurance

Most of the countries participating in the ICP Forests programme used the recommended methods for digestion, ashing and analysis as described in the manual on sampling and analysis of needles and leaves (Rautio *et al.*, 2010; Rautio

& Fu¨ rst, 2013). Other methods were allowed; they were validated by comparing the element concentrations obtained with these methods to those of reference standard samples.

A good data quality is the basis for detecting small trends in nature (Sulkava *et al.*, 2007). Therefore, quality assurance procedures (e.g., plausible ranges, control charts) were used by each country to check data before submission; these proce- dures are described in the manual on Quality Assurance and Control in Laboratories (Ko€nig *et al.*, 2013). The national labo- ratories also participate in regular needle/leaf interlaboratory testing organised by the Forest Foliar Coordinating Centre of

where *b* and *a* are respectively the slope and the intercept obtained from the data using Eqn (1). The normality of the foliar variable was evaluated based on the skewness and the kurtosis whose values must be between -2 and 2 to accept the normality of the distribution (Rupert & Miller, 1986). For some variables, these criteria were not satisfied. In these cases, the values of the foliar variable were log-transformed to improve the normality of the distribution. When log-transformed values were used, relative change in 10 years was calculated according to back-transformed values.

10*a*þ5·*b* - 10*a*-5·*b*

ICP Forests (Fu¨ rst, 2014). Depending on the variable (N, S, P,

Ca, Mg, K), the tolerable limits in the interlaboratory compari-

relative change in 10 years ð%Þ ¼

10*a* · 100 ð3Þ

son are allowed to vary from 10% to 20% of the mean (see details in Rautio *et al.*, 2010; Fu¨ rst, 2014).

## Statistical analyses

When individual chemical analyses and mass measurements were performed for each sampled tree, a mean value per plot, species and needle class was calculated.

To account for the complex correlation structure of the data- set, linear mixed models were used to detect the temporal trends in foliar nutrition at the European scale. Foliar nutrition was characterised by several variables [fol. var. in Eqn (1)]: dry mass of 100 leaves or 1000 needles, foliar N, P, S, Ca, Mg and K concentrations and contents and several foliar ratios (N/P, N/Mg, N/K, N/S, Ca/K). The content of a given nutri- ent in 100 leaves or 1000 needles was calculated by multiply- ing its concentration by its leaf or needle mass. A linear mixed model was fitted for each species, needle class (coniferous spe- cies) and foliar variable. As an intercept corresponding to the year 0 has no biological meaning, the variable describing the time was centred on the year 2000 (which is more or less the middle of the monitoring period). Centring the independent variable provides an intercept and a slope independent from each other. The intercept and the slope of the linear models contain both fixed and random components. For the intercept, the parameter *a* is the fixed part (which can be interpreted as an estimate of the mean mass, concentration, content or ratio in 2000) and ctryint and plotint are two random factors describ- ing the country and plot effects, respectively. For the slope, the parameter *b* is the fixed part and plotslp is a random coeffi- cient that accounts for slope random variation between plots [Eqn. (1)].

The fixed part of Eqn (1) [Eqn. (4)] was also fitted plot by

plot for all foliar nutrient concentrations to analyse the rela- tionships between the estimated mean foliar concentrations in 2000 and the relative changes in concentrations.

fol. var. ¼ *a* þ *b* · ðyear - 2000Þþ eð0; r2 Þ ð4Þ

Only the plots with more than four sampling dates covering at least a 10-year period were retained for this approach to get more precise estimates of a and b.

The fitting of Eqns (1) and (4) was carried out with the MIXED procedure of the SAS software (version 9.3; SAS insti- tute Inc., Cary, NC, USA).

# Results

## Foliar mass

From 1992 to 2009, the mass of 1000 needles signifi- cantly rose in current-year needles of *P. abies* (9.7% in 10 years) while the decreasing trends observed for both needle classes of *A. alba* were not significant (Fig. 2). The mass of 100 leaves increased for all broadleaved species but this increase was significant only in *F. sylv- atica* (7.6% in 10 years; Fig. 3).

## Nitrogen

The foliar N status was quite high for most broadleaved plots while it was low or deficient in a number of conif- erous plots (66% of the *P. abies* plots and 35% of the

1. *sylvestris* plots; Fig. 4). During the monitoring period, foliar N concentration significantly decreased in the

2 2

fol. var. ¼ ½*a* þ ctryintð0; rci Þþ plotintð0; rpi Þ] þ ½*b*

2 2

þ plotslpð0; rps Þ] · ðyear - 2000Þþ eð0; r Þ ð1Þ

where r2 , r2 , r2 , and r2 are the variances of the random fac-

current-year foliage (leaves or needles) of *F. sylvatica*,

1. *petraea* and *A. alba* and in 1-year-old needles of *P.*

*abies*. The N content in 100 leaves or 1000 needles signif- icantly increased for *F. sylvatica* and *P. abies* (current-

ci pi ps

tors ‘country’ and ‘plot’, of the random coefficient ‘plot’ and

of the residual term (e), respectively.

For each foliar parameter, the fitted values of the fixed parameters were then used to calculate the relative change in 10 years according to the following equation:

*b* · 10

year needles) and significantly decreased for *A. alba*

(current-year needles) (Figs 2 and 3).

## Phosphorus

Depending on the tree species, the foliar P status was

relative change in 10 years ð%Þ ¼

*a* · 100 ð2Þ

low or deficient in a proportion of plots ranging from

15 *)* 15

Relative change

for a 10-year period (%)

Relative change

for a 10-year period (%)

|  |  |  |  |
| --- | --- | --- | --- |
| *Abies alba (current year needles*  **\***  NS  NS NS | | | |
| **\***  NS | NS |  | NS NS |
|  | |
| **\***  **\*** | |

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| *Abies alba (1-year-old needles)*  NS  NS | | | | |
| NS | | | | NS |
| NS |  | NS | NS NS | |
| NS  **\***  **\*** | | NS N | | |

10 10

5 5

0

–5

–10

–15

0

NS –5 S

NS

–10

–15

Mass N P S Ca Mg K Mass N P**(\*)**

S Ca Mg K

15 15

**\***

*Picea abies (current year needles)*

**\***

**\***

**\***

**\***

**(\*)**

**(\*)**

**(\*)**

NS

NS

NS

**\***

**\***

*Picea abies (1-year-old needles)*

**(\*)** NS

NS

NS

**\*** NS

NS

NS

**\***

**(\*)**

**\***

**\* \***

Relative change

for a 10-year period (%)

Relative change

for a 10-year period (%)

10 10

5 5

–5

–10

–5

–10

–15

–15

Mass N P S Ca Mg K Mass N P S Ca Mg K

15 15

Relative change

for a 10-year period (%)

Relative change

for a 10-year period (%)

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| *Pinus sylvestris (current year needles)*  NS  NS NSNS NS NS | | | | |
|  |  | | NS NS | NS |
| **\*** NS | | NS  **\*** | | |

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| *Pinus sylvestris (1-year-old needles)*  **(\*)** | | | | |
| NS | | **\*** | NS | NSNS |
| NS | NSNS | NS |
| NS | | NS  NS | | |

10 10

5 5

0 0

–5 –5

–10

–10

–15

–15

Mass N P S Ca Mg K Mass N P S Ca Mg K

Concentrations Contents

Fig. 2 Relative changes in needle mass and in nutrient concentrations and contents of coniferous species for a 10-year period. Signifi- cant changes are indicated with \* when *P* < 0.05 and with (\*) when *P <* 0.1.

22% (*P. sylvestris*) to 74% (*Q. petraea*) (Fig. 4). Foliar P concentration decreased over time in all cases except for the 1-year-old needles of *P. sylvestris* but this decreasing trend was only significant for the current- year leaves or needles of *F. sylvatica*, *Q. petraea* and

*P. sylvestris* and for the 1-year-old needles of *A. alba* and

*P. abies*. The P content in 100 leaves or 1000 needles significantly decreased for the current-year needles of

*A. alba* while it increased for the current-year needles of

*P. abies* (Figs 2 and 3).

## Sulphur

The foliar S status was at a medium level for most of the broadleaved plots but it was low for 88% of the

*P. abies* plots and 68% of the *P. sylvestris* plots (Fig. 4). Foliar S concentration decreased over time in all cases, except for the current-year needles of *A. alba*; this decrease was significant for the current-year foliage of *F. sylvatica*, *Q. petraea* and *P. sylvestris* and for both needle age classes of *P. abies*. The S content

in 100 leaves significantly increased in *F. sylvatica*

(Figs 2 and 3).

## Calcium

The foliar Ca status was low or deficient in about one-third of the plots with some variations among tree species (from 18% to 40%). Foliar Ca concentration significantly decreased for *F. sylvatica* and *Q. petraea* while it significantly increased for the 1-year-old needles of *P. sylvestris*. The Ca content in 1000 needles significantly increased for the current-year needles of

*P. abies* (Figs 2 and 3).

## Magnesium

The foliar Mg status was low or deficient in <20% of the plots for all tree species, except *F. sylvatica* (29%; Fig. 4). Foliar Mg concentration significantly decreased for

*F. sylvatica* while it significantly increased for the current-year needles of *A. alba*. The Mg content in 1000

15

10

Relative change

for a 10-year period (%)

5

0

–5

–10

ratio significantly increased in *F. sylvatica* and in cur- rent-year needles of *P. abies* and *P. sylvestris.* The foliar N to Mg ratio significantly decreased in current-year needles of *A. alba* and *P. abies*. The foliar N to K ratio significantly increased in *Q. petraea* and in 1-year-old needles of *P. abies*. The foliar Ca to K ratio significantly increased in 1-year-old needles of the coniferous spe- cies (Table 1).

–15

15

10

Relative change

for a 10-year period (%)

5

0

–5

–10

–15

15

10

Relative change

for a 10-year period (%)

5

0

–5

–10

–15

Mass N P S Ca Mg K

*Fagus sylvatica*

**\***

NS

**\***

**\***

NS

NS

**\***

NS

NS

**\***

**\***

**\***

**\***

NS

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| NS |  | NS | *Quercus petraea*  NS NS | | |
| **\*** | | | | NS  **\*** NS | |
| **(\*) \***  **\*** | | | | | **\*** |

Mass N P S Ca Mg K

**(\*)**

NS

*Quercus robur*

NS

NS

NS

NS NS

NS

NS

NS

NS

NS

NS

Mass N P S Ca Mg K

Concentrations Contents

## Temporal trends at the plot level

Based on the results of the model fitting [Eqn (4)] car- ried out for each plot individually, we analysed the relationships between the estimated mean foliar con- centrations in 2000 [parameter *a* in Eqn (4)] and the rel- ative changes in foliar concentrations [calculated based on Eqn (2)]. In *Q. Petraea* and *P. sylvestris*, a positive effect of the mean foliar P concentration was observed on the relative change in foliar P concentration, in other words the relative change (which was negative for the majority of the plots) was the smaller the higher was the estimated P concentrations in 2000 (Fig. 5). For the same tree species, the mean foliar S concentration had a negative effect on the relative change in foliar S concen- tration; the decrease in foliar S concentration was more pronounced in plots exhibiting a higher estimated foliar S concentration in 2000 (Fig. 5). For Mg, the effect of the mean foliar concentration on the relative change in concentration was also negative in *F. sylvatica* and *P. abies*. In *P. abies*, the positive change in foliar Mg concentration decreased with the mean foliar Mg con-

Fig. 3 Relative changes in leaf mass and in nutrient concentra- tions and contents of broadleaved species for a 10-year period. Significant changes are indicated with \* when *P* < 0.05 and with (\*) when *P <* 0.1.

needles significantly increased for the current-year needles of *P. abies* (Figs 2 and 3).

## Potassium

The foliar K status was low or deficient in less than 20% of the plots for all tree species, except *P. abies* (36%; Fig. 4). Foliar K concentration significantly decreased for *Q. petraea* and for *P. abies* (both needle classes). The K content in 1000 needles significantly decreased for the 1-year-old needles of *P. abies* (Figs 2 and 3).

## Foliar ratios

The foliar N to P ratio significantly increased in *F. sylv- atica*, *Q. petraea*, 1-year-old needles of *P. abies* and in current-year needles of *P. sylvestris*. The foliar N to S

centration and in *F. sylvatica*, the negative change in foliar Mg concentration was on average more pro- nounced in plots with a better foliar Mg status (Fig. 5).

# Discussion

## Identifying potentially limiting nutrients

Potentially growth-limiting nutrients can be identified by analysing the proportions of plots presenting differ- ent foliar nutrient statuses (Fig. 4). In broadleaved spe- cies, P was the main limiting nutrient while *P. abies* and

*P. sylvestris* were colimited by N and P. To a lesser extent, base cations were also limiting in a number of broadleaved and coniferous plots. According to the classification values fixed by the Foliar Expert Panel of ICP Forests (Stefan *et al.*, 1997), most of the *P. abies* and

*P. sylvestris* had low foliar S concentration.

According to the thresholds set by Mellert & Go€ttlein (2012), N deficiency was not detected in the most N demanding species (broadleaved species). In the cases of *P. abies* and *P. sylvestris,* whose plots are partly located in regions with low N deposition (e.g.,

100

80

Number of plots

60

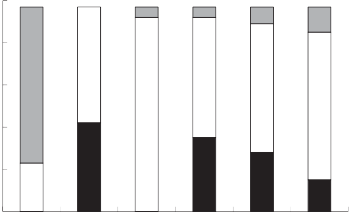
40

20

0

*Fagus sylvatica*

35



30

Number of plots

25

20

15

10

5

0

*Quercus petraea*

N P S Ca Mg K N P S Ca Mg K

35

30

Number of plots

25

20

15

10

5

0

100

80

Number of plots

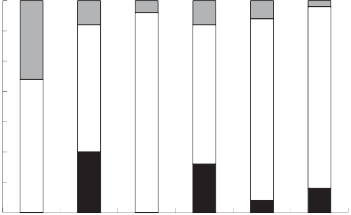
60

40

20

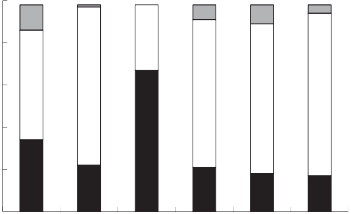
0

*Quercus robur*



N P S Ca Mg K

*Pinus sylvestris*



N P S Ca Mg K

140

120

Number of plots

100

80

60

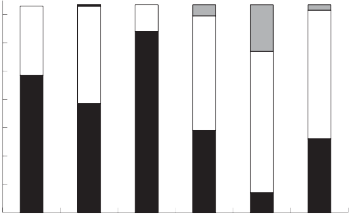
40

20

0

*Picea abies*

N P S Ca Mg K



Surplus (High to Toxic) Normal (Medium) Deficiency (Low)

Fig. 4 Number of plots in the three foliar nutrient status classes. Each bar presents the number of plots in which the mean foliar con- centration of the current-year leaves or needles falls within the thresholds set by Mellert & Go€ttlein (2012) for N, P, Ca, Mg and K and by Stefan *et al.* (1997) for S (in parenthesis).

Scandinavia, see Mustaja¨ rvi *et al.*, 2008), large portion of the plots were classified as N deficient. In these regions, N is generally limiting tree growth as sug- gested by the positive effect of N fertilisation on stem volume increment in several N fertilisation experiments in boreal forests in the Nordic countries (Ingerslev *et al.*, 2001; Ho€gberg *et al.*, 2006). In contrast, the spatial distribution of broadleaved plots is more centred on Central Europe where N deposition is higher (Waldner *et al*., 2014). In these N enriched forests, N is no longer limiting but other nutrients such as P can become limit- ing as seems to be the case in broadleaved species here. These results are in agreement with several studies. Based on 135 permanent observation plots distributed across Switzerland and on seven N addition experi- ments, Braun *et al.* (2010) showed that Swiss forests (*P. abies* and *F. sylvatica*) were saturated in N and that this N saturation induced a P limitation. In Germany, P limitation was reported by Mellert & Go€ttlein (2013) for

*P. abies* stands and by Prietzel *et al.* (2008) for *P. sylves- tris* stands. However, Mellert & Ewald (2014) observed persisting N limitation in the *P. abies* stands of Bavarian Alps despite considerable deposition inputs; this was

attributed to a high rate of N retention in the organic horizons.

## Decreasing temporal trends in foliar nutrient concentrations

Among the 22 significant temporal trends in foliar nutrient concentrations detected in this study, 20 are decreasing and 2 are increasing (Figs 2 and 3). The rela- tive change in foliar concentrations for a 10-year period ranges from -11% to 7.5%. Such temporal trends in foliar concentrations have previously been observed regionally but not on a European scale.

In Switzerland, Flu¨ ckiger & Braun (1998) reported an increase in foliar N concentration in *F. sylvatica* and a decrease in foliar P concentration in *F. sylvatica* and

*P. abies* based on foliar analyses carried out from 1984 to 1995. Braun *et al.* (2010) reanalysed this foliar dataset with a time series extended until 2007, and found a sig- nificant decrease in N and P foliar concentrations for both *F. sylvatica* and *P. abies*. Twenty-five years after the first foliage sampling, Duquesnay *et al.* (2000) resam- pled 118 *F. sylvatica* stands in North-Eastern France and

Table 1 Estimates of the mean foliar ratios in 2000 and of the relative changes in foliar ratios for a 10-year period. Leaf age: 0 rep- resents current-year leaves/needles and 1 represents 1-year-old needles. The underlined and bold values are significant at *P* < 0.05 while the values in bold (not underlined) are significant at *P* < 0.1

Tree species Leaf age N/P N/S N/Mg N/K Ca/K

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Estimate of the mean foliar ratio in 2000 [parameter a of Eqn (1)] | | | | | | |
| *Fagus sylvatica* | 0 | 20.31 | 14.78 | 18.81 | 3.37 | 1.08 |
| *Quercus petraea* | 0 | 18.93 | 14.93 | 15.51 | 3.01 | 0.90 |
| *Quercus robur* | 0 | 16.35 | 14.69 | 15.29 | 3.02 | 0.78 |
| *Abies alba* | 0 | 10.86 | 11.66 | 9.47 | 2.64 | 1.18 |
|  | 1 | 12.67 | 11.36 | 14.10 | 3.03 | 1.99 |
| *Picea abies* | 0 | 9.49 | 14.30 | 12.56 | 2.47 | 0.69 |
|  | 1 | 10.77 | 13.57 | 13.86 | 2.60 | 1.06 |
| *Pinus sylvestris* | 0 | 11.09 | 14.45 | 15.96 | 2.91 | 0.51 |
|  | 1 | 12.27 | 15.02 | 19.32 | 3.23 | 0.91 |
| Estimate of the relative change in foliar ratio for a 10-year period [Eqns (2) or (3)] | | | | | | |
| *Fagus sylvatica* | 0 | 3.9 | 2.5 | 4.3 | -0.3 | -1.4 |
| *Quercus petraea* | 0 | 8.2 | -0.4 | -1.3 | 5.7 | 4.0 |
| *Quercus robur* | 0 | 4.1 | 0.9 | 0.5 | 1.4 | 0.2 |
| *Abies alba* | 0 | 4.2 | -5.1 | - 10.4 | -0.7 | 3.2 |
|  | 1 | 9.4 | -3.0 | -5.1 | 2.7 | 12.2 |
| *Picea abies* | 0 | -0.3 | 3.2 | - 4.2 | 2.3 | 3.5 |
|  | 1 | 3.0 | 2.2 | -2.6 | 3.3 | 9.5 |
| *Pinus sylvestris* | 0 | 3.4 | 5.2 | -2.6 | -2.5 | -4.0 |
|  | 1 | 1.5 | -1.2 | 1.3 | 0.0 | 6.3 |

observed an increase in foliar N concentration and a decrease in foliar P, Ca and Mg concentrations. By anal- ysing the foliar data of the French, Walloon and Lux- embourg broadleaved plots of ICP Forests, Jonard *et al.* (2009) detected a general decrease in P concentration between 1993 and 2005. Except for the increasing trend in foliar N concentration (*F. sylvatica*), these results for broadleaved species are in accordance with the tempo- ral changes highlighted in this study.

Based on foliar samples collected in 42 *P. abies* and *P. sylvestris* stands in Southern Sweden from 1985 to 1994, Thelin *et al.* (1998) observed decreasing trends in foliar K concentration in both species as well as in foliar S concentration for *P. sylvestris*. Increasing trends in foliar Ca and Mg concentrations in *P. abies* were also seen. These trends are consistent with our results except that we did not observe any trends in foliar K concentration for *P. sylvestris*. Using foliar data from control plots of fertilisation trials conducted in *P. abies* and *P. sylvestris* stands of Central Europe and Scandinavia since 1960, Mellert *et al.* (2004) also detected mainly decreasing trends for macronutrients, except for foliar N concen- tration of *P. sylvestris* which increased in Central Eur- ope. In Austria a clear decrease in S in foliage have also been observed (Fu¨ rst *et al.*, 2003). In this study (Aus- trian Bio-Indicator Grid) 760 *P. abies* and *P. sylvestris* stands where collected and analysed annually from 1983 to 2000.

All the temporal trends in foliar nutrient concentra- tions detected in this study are generally consistent with the trends reported in earlier regional studies. When monitoring the same sample plots (and same sample trees) for a long period of time, one cannot avoid the fact that the temporal trends in foliar nutri- tion are affected by the age of the sample trees. How- ever, ageing can only explain relatively small changes (about 1% in 10 years according to Braun *et al.*, 2010) while the magnitude of the trends observed here are much higher. Foliar nutrient concentrations are also influenced by interannual variations in meteorological conditions (Stefan & Gabler, 1998; Potocˇi'c *et al.*, 2005; Jonard *et al.*, 2009). Drought periods decrease nutrient availability in the soil by reducing organic matter min- eralisation and ion mobility and affect the nutrient uptake capacity by modifying carbon allocation to roots, root absorbing surface and maximum absorption rates (Kreuzwieser & Gessler, 2010). Thus more fre- quent drought periods and heat waves observed in Europe during the last decades (Beniston & Diaz, 2004) or increasing insect attacks in the canopy (Pitman *et al.*, 2010) might also have played a role in the long-term changes in foliar chemistry seen here.

The significant trends in foliar N/nutrient ratios were positive for P, S and K and negative for Mg indicating that P, S and K foliar nutrition was deteriorating in comparison to N nutrition while Mg nutrition was

50

Relative change in foliar P concentration (%)

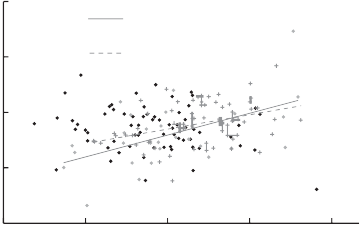
25

0

–25

–50

*Q. petraea*: *y* = –53.06 + 32.91x



*P* = 0.0028, *R*² = 0.29

*P. sylvestris*: *y* = –35.63 + 21.59x

*P* = 0.0005, *R*² = 0.15

diminished. We found that the decrease in foliar S con- centration was more pronounced in plots exhibiting high foliar S concentration (Fig. 5). Since, these plots are most likely those that have received higher S depo- sition in the past, reduced S deposition is mirrored more clearly on foliar S nutrition in these plots. High N deposition (occurring mainly in Central Europe) can affect P nutrition by at least two means. Firstly, N depo-

0.7 1.0 1.3 1.6 1.9

Estimated foliar P in 2000 (mg g–1)

sition acts as a fertiliser increasing tree growth, which increases need for other nutrients such as P (Pen~uelas

50

Relative change in foliar S concentration (%)

25

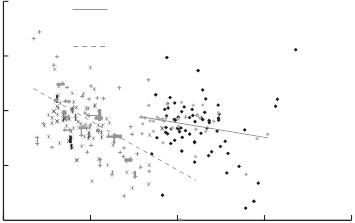
0

–25

–50

*Q. petraea*: *y* = 14.25 – 13.19x

*P* = 0.0552, *R*² = 0.13



*P. sylvestris*: *y* = 40.44 – 45.01x

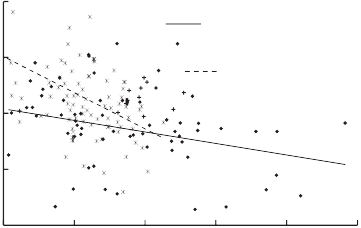
*P* = <0.0001, *R*² = 0.34

*et al.*, 2012). Secondly, increased N availability reduces fine root biomass and negatively affects the develop- ment of mycorrhiza (Nilsson & Wallander, 2003; Kjøller *et al.*, 2012), which in turn plays a very important role in the P uptake of trees (Wallander, 2000). Also S depo- sition can have an effect on P nutrition. Since sulphate and phosphate are partly adsorbed on the same anion

0.5 1.0 1.5 2.0 2.5

Estimated foliar S in 2000 (mg g–1)

50



*F. sylvatica*: *y* = 7.16 – 10.30x

*P* = 0.0213, *R*² = 0.08

*P. abies*: *y* = 41.58 – 32.34x

*P* = 0.0002, *R*² = 0.16

Relative change in foliar Mg concentration (%)

25

0

–25

–50

0.5 1.0 1.5 2.0 2.5 3.0

Estimated foliar Mg in 2000 (mg g–1)

F. sylvatica Q. Petraea Q. robur

A. alba P. abies P.sylvestris

Fig. 5 Relationships between the relative change in concentra- tion [Eqn (2)] and the estimated foliar concentration in 2000 [parameter *a* in Eqn (4)]. Only tree species exhibiting a signifi- cant trend in foliar concentration (Figs 2 and 3) are represented. When the relationship is significant, the regression line is pre- sented as well as the equation, *P* value and *R*².

improving. Foliar Ca to K ratio showed an increasing trend in the 1-year-old needles of the coniferous spe- cies, suggesting either increased soil Ca availability or problems in K nutrition.

The positive time trends in foliar N to P, S and K ratios is, at least partly due to continuing high N depo- sition in many European forest ecosystems (Posch *et al.*, 2012) despite the decrease in N deposition observed since the 1980s (Fischer *et al.*, 2007; Lorenz *et al.*, 2008). In the case of foliar N to S ratio, the positive temporal trend are mainly due to the sharper decline in S than in N deposition, which has reduced the sulphate concen- tration in the soil solution (Jonard *et al.*, 2012) and led to lower S uptake from soil. Furthermore, due to decreased S deposition, direct foliar uptake of S has

exchangers of the soil constituents, the decrease in sul-

phate concentration in the soil solution could have favoured a higher adsorption of phosphate, therefore reducing its availability for trees (Geelhoed *et al.*, 1997). In *Q. petraea* and *P. sylvestris*, the negative changes in foliar P concentration were more pronounced in plots with a poorer foliar P status (Fig. 5). These observations support the hypothesis of an increased P demand due to higher tree productivity. In the plots with the lower soil P availability, the increased P requirement cannot be satisfied.

The negative temporal trend in foliar N to Mg ratio could be an indication that some forest ecosystems are progressively recovering from past acidification due to the sharp decline in acidifying deposition observed in Europe (Fischer *et al.*, 2007; Lorenz *et al.*, 2008). In *P. abies*, the positive change in foliar Mg concentration was more marked in plots with a low foliar Mg status. This is in agreement with the find- ings of Cools & De Vos (2011) who compared two soil inventories carried out in 1985–1996 and in 2006– 2008, and found an increase in soil pH and in base saturation in soils with low pH (pH CaCl2 < 4) and low soil buffering capacity (BS < 20%). In *F. sylvatica*, the negative changes in foliar Mg concentration were more pronounced in plots with a better foliar Mg sta- tus (Fig. 5). These plots probably continue to acidify (Cools & De Vos, 2011), possibly due to N leaching in N saturated ecosystems (many *F. sylvatica* plots with a surplus N nutrition, see Fig. 4) or due to nutrient removal associated with timber harvesting. In the monitoring plots studied here, nutrient export could have contributed to reduced soil nutrient avail- ability but this effect was probably limited during the study period since only light, selective cuttings, if any, were carried out and no plots with clear cuttings were included here.

At present, there is a growing demand for bioenergy in Europe, and it is therefore important to highlight that timber harvesting also contributes to soil acidification. Intensified wood biomass harvesting to satisfy the demand for renewable energy increase nutrient removal from forests if small branches, twigs, foliage, and roots are used as bioenergy since these tree compo- nents have higher nutrient concentrations than the stem and coarse branches (Andre´ *et al.*, 2010; Merila¨ *et al.*, 2014).

## Increase in foliar mass and dilution effect

Analysing the temporal changes in the mass of 100 leaves or 1000 needles is essential to understand the time trends in foliar nutrient concentrations. An increase in foliar mass can reduce foliar nutrient con- centrations by dilution while a decrease in foliar mass can lead to an increase in foliar nutrient concentrations. For broadleaved species, upward trends in foliar mass were observed but this temporal change was significant only in *F. sylvatica* (Fig. 3). The only significant tempo- ral trend in conifers was seen in *P. abies* in which mass of current-year needles increased by 10% in 10 years (Fig. 2). This increase is quite substantial since increases in foliage mass of this magnitude are usually seen in fertilisation experiments (Maier *et al.*, 2008; Krause *et al.*, 2012). In many parts of Europe elevated N depo- sition, originating from numerous anthropogenic sources (fossil fuels, agriculture etc.), functions as fertil- iser that can enhance tree productivity, including increased foliage growth. In broadleaved species in which foliar N was at an adequate level, the increase in foliage mass can be further enhanced by the global enrichment in atmospheric CO2 as well as the rise in air temperature and prolonged vegetation period (Boisve- nue & Running, 2006). The increase in productivity could also be explained by the development stage of the forests if most of them were in phase of exponential growth during the study period. Indeed, the periodic annual increment in volume of a forest stand increases to a maximum value as tree matures and then slowly declines during the rest of the silvicultural cycle. Depending on the site index, the maximum periodic increment occurs when trees are between 20 and 50 years old in case of the coniferous species studied here and between 40 and 75 years in case of the studied broadleaved species (INRA, ONF, ENGREF, 1984; Da- gnelie *et al.*, 1988). At the beginning of the study period, the coniferous species on our sample plots were on average 60–80 years old and 80–100 years old in case of broadleaved species. The development stage of the studied forest stands can therefore not explain the increase in productivity since the majority of these

stands had passed the period of maximum growth already before the beginning of the study period.

## Temporal trends in nutrient contents

Increased foliar mass observed in *F. sylvatica*, *Q. robur* and *P. abies* (current-year needles) was accompanied by a near systematic increase in nutrient contents, except for the foliar Mg content of *F. sylvatica* (Fig. 3). In *P. abies,* the increase in the nutrient contents of current- year needles seems to have occurred at the expense of the nutrient contents of older needles. Part of the increased nutrient demand (increased foliar mass) of *P. abies* was probably met by a higher nutrient retransloca- tion from 1-year-old needles (Wyttenbach *et al.*, 1995; Bauer *et al.*, 1997) whose nutrient contents decreased with time. This was the case in all major nutrients except in Ca, which is a rather phloem immobile nutri- ent (Marschner, 2012).

Although the nutrient contents of current-year leaves or needles rose for *F. sylvatica*, *Q. robur* and *P. abies*, their increased nutrient demand associated with their increased growth remained partly unsatisfied by soil nutrient supply leading to a decrease in nutrient con- centration (dilution effect).

## Alarming trends in foliar nutrition

Among the temporal changes in foliar nutrition observed in this study, some trends demand immediate notice. A trend is considered to be alarming when a decrease in foliar concentration was observed for a lim- iting nutrient. According to this criterion, we found a significant deterioration in P nutrition in *F. sylvatica*, *Q. Petraea* and *P. sylvestris.* In *Q. Petraea* and *P. sylvestris*, the decrease in foliar P concentration was more pro- nounced on plots with low foliar P status, meaning that trees with latent P deficiency could become deficient in the near future. Regarding *A. alba* and *P. abies*, no sig- nificant trends in foliar P concentration were observed in current-year needles while decreasing trends were noticed in 1-year-old needles, possibly indicating an increased P retranslocation from older needles in response to increased P requirement. This deterioration in P nutrition will certainly limit tree response to increased atmospheric CO2 as well as the C sequestra- tion capacity of European forests. It could also induce tree health problems since the N to P ratio of all tree species (Table 1) was clearly above the thresholds beyond which defoliation has been found to increase in broadleaved (14.8) and coniferous (7.3) stands (Vere- soglou *et al.*, 2014). These results are totally consistent with several recent global studies showing that the unbalanced inputs of C and N relative to P induced

significant changes in organism stoichiometry resulting in profound and uncertain consequences on the struc- ture, functioning and diversity of terrestrial and aquatic ecosystems (Pen~uelas *et al.*, 2012, 2013 and Sardans *et al.*, 2012). Among others, Pen~uelas *et al.* (2013) showed that the limited P availability is likely to reduce carbon storage in natural ecosystems during the course of the century. Based on a synthesis study of 92 forests in different climate zones, Fernandez-Martinez *et al.* (2014) observed that only forests with adequate nutri- ent supply are able to augment their net ecosystem pro- ductivity with increasing gross primary production.

While no broadleaved plots were found to have low foliar S status, high proportions of the *P. abies* and *P. sylvestris* plots (88% and 68%, respectively) exhibited low foliar S concentrations in current-year needles. A decrease in foliar S concentration was observed in current-year needles of both coniferous species and for 1-year-old needles of *P. abies*. These negative trends in foliar S concentrations seem how- ever less alarming than for P since the decrease was more pronounced in plots with higher foliar S status (Fig. 5) and since the mean N to S ratio (Table 1) is not indicating any deficiency in S (Bonneau, 1995). In all species, the mean N to S ratio is equal or below 15 which corresponds to the level needed to produce an optimal level of proteins for tree growth and maintenance (Bonneau, 1995). The downward trends in foliar S concentration should however be moni- tored because the current trend could eventually lead to S deficiency in the future.

In *F. sylvatica*, the foliar Mg status (which was esti- mated to be low or deficient in 29% of the plots in 2000) significantly decreased during the 1992–2009 period. This decline in foliar Mg concentration was however more marked in plots with a high foliar Mg status. Regarding K, a decreasing trend in foliar concentration was observed in *Q. petraea* and for *P. abies* which had respectively 12% and 35% of plots with low or deficient foliar K status. For Ca, we also notice a decrease in foliar concentration of *F. sylvatica* and *Q. petraea* (36% and 18% of plots with low foliar Ca status, respectively).

## Concluding remarks

Altogether our results show a clear deterioration in P nutrition during the past two decades in some of the main tree species. Our study also highlights some downward trends that should be monitored closely in the future since they could become alarming: e.g., decrease in foliar S concentration in *P. abies* and *P. syl- vestris*, in foliar Mg concentration in *F. sylvatica*, in foliar K concentration in *Q. petraea* and *P. abies* and in foliar

Ca concentration in *F. sylvatica* and *Q. petraea*. As tree nutrient status exerts a tight control on net ecosystem productivity, this deterioration in tree nutrition could have a strong impact on the response of forest ecosys- tems to climate change. Nutrient availability and tree nutrition should therefore be accounted for in global carbon cycle to allow climate models to better predict C sequestration capacity of forests and avoid overestimations.

In Europe, increased tree productivity likely due to high N deposition and also possibly to global atmo- spheric CO2 enrichment has led to a higher nutrient demand by trees. As the soil nutrient supply was prob- ably not always sufficient to meet the growing demands by trees, this could partly explain the deterio- ration of tree mineral nutrition. In addition, the sharp decline in S deposition has significantly reduced its availability, while elevated N deposition could have negatively affected nutrient uptake capacity of trees and lead to nutrient imbalances.

This study also highlights the importance of long- term forest monitoring in following spatiotemporal trends in forest ecosystems. Now that Europe is dra- matically increasing its wood biomass harvesting to satisfy the demands for bioenergy, it is crucial to take into account nutrient limitations for forest growth to avoid overestimations of forest productivity in response to elevated atmospheric CO2 when develop- ing global climate models. To be able to monitor changes in carbon sequestration capacity of forests, and the underlying factors like tree nutrition affecting this capacity, a European wide monitoring network is needed which is able to detect and act on alarming trends as they appear.

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