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ORIGINAL PAPER

# Exceedance of critical loads and of critical limits impacts tree nutrition across Europe

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**Abstract**
· ***Key message*** **Exceedance of critical limits in soil solution samples was more frequent in intensively monitored forest plots across Europe with critical loads for acidity and eutrophication exceeded compared to other plots from the same network. Elevated inorganic nitrogen concentrations in soil solution tended to be related to less favourable nutritional status**.

· ***Context*** Forests have been exposed to elevated atmospheric deposition of acidifying and eutrophying sulphur and nitrogen compounds for decades. Critical loads have been identified, below which damage due to acidification and eutrophication are not expected to occur.
· ***Aims*** We explored the relationship between the exceedance of critical loads and inorganic nitrogen concentration, the base cation to aluminium ratio in soil solutions, as well as the nutritional status of trees.
· ***Methods*** We used recent data describing deposition, elemental concentrations in soil solution and foliage, as well as the level of damage to foliage recorded at forest plots of the ICP Forests intensive monitoring network across Europe.
· ***Results*** Critical loads for inorganic nitrogen deposition were exceeded on about a third to half of the forest plots. Elevated

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inorganic nitrogen concentrations in soil solution occurred more frequently among these plots. Indications of nutrient imbalances, such as low magnesium concentration in foliage or discolouration of needles and leaves, were seldom but appeared more frequently on plots where the critical limits for soil solution were exceeded.
• ***Conclusion*** The findings support the hypothesis that elevated nitrogen and sulphur deposition can lead to imbalances in tree nutrition.

**Keywords** Inorganic nitrogen concentration in soil solution · Base cation to aluminium ratio · Tree nutrition · Foliage · ICP Forests

## 1 Introduction

Forest ecosystems have been exposed to elevated atmospheric deposition of sulphur (S) and nitrogen (N), mainly as sulphate ($SO_4^{2-}$) and inorganic N, for more than five decades. The main reason being a large increase in the anthropogenic emissions of N and S compounds in the second half of the last century. The elevated deposition of N and S affects forest ecosystems through several processes.

Inorganic N and $SO_4^{2-}$ deposition may accelerate acidification of forest soils through leaching of strong acid anions, mainly nitrate ($NO_3^-$) and $SO_4^{2-}$, accompanied by base cations (BC) such as calcium ($Ca^{2+}$), magnesium ($Mg^{2+}$), and potassium ($K^+$) (Ulrich et al. 1980). Soil acidification may result in (i) depletion of soil BC and (ii) mobilisation of aluminium ($Al^{3+}$) into soil solution, possibly with adverse effects on fine roots and associated mycorrhizal fungi (e.g. de Wit et al. 2010). Toxic effects of dissolved Al are reduced by the presence of dissolved base cations. The molar ratio *Bc/Al* (molc molc$^{-1}$) in soil solution, where *Bc* is the sum of the molar concentrations of the base cations $Ca^{2+}$, $Mg^{2+}$ and $K^+$, and *Al* that of dissolved aluminium species in soil solution, has been suggested as a criterion to assess Al toxicity (Cronan and Grigal 1995).

Enhanced N supply may stimulate tree growth in N limited stands. In excess, however, N may induce (i) nutrient imbalances; (ii) increased sensitivity to frost, insects, and fungi; and (iii) elevated $NO_3^-$ and ammonium ($NH_4^+$) leaching from the root zone (Aber et al. 1989). The concentration of inorganic N in soil

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solution ($Nmin = NO_3^- + NH_4^+$, mg $L^{-1}$), $NO_3^-$ leaching, the nutritional status of trees, as well as the organic carbon (C) to N ratio in the forest floor (*C/N*, kg $kg^{-1}$) have all been suggested as indicators of the N saturation status of forests (Gundersen et al. 2006; Dise et al. 2009).

In the frame of the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP), long-term effects of atmospheric deposition on ecosystems are generally assessed based on the concepts of critical loads and critical limits. Such critical loads and critical limits are defined as quantitative estimates of an exposure to deposition loads or levels below which significant harmful effects on specified sensitive elements of the environment do not occur according to current knowledge (Nilsson and Grennfelt 1988).

Two approaches are widely used for estimating critical loads for atmospheric N deposition. The first approach is to compile empirical observations to provide a range of typical critical loads for N deposition for each ecosystem type, e.g. 5 to 15 kg N $ha^{-1}$ $year^{-1}$ for coniferous woodland and 10 to 20 kg N $ha^{-1}$ $year^{-1}$ for broadleaved deciduous woodland (empirical critical loads for N deposition) (Bobbink and Hettelingh 2011). The second approach derives critical loads for N deposition from a criterion applied to nutrient fluxes or levels in an ecosystem model and is generally implemented with a steady-state mass balance (SSMB) of input sources and output sinks (Sverdrup and de Vries 1994). Pools are excluded and assumed to be irrelevant under long-term considerations. A common criterion for calculating critical loads for N deposition using the SSMB is that the leaching flux of N below the root zone should not exceed an acceptable level (Spranger et al. 2004). This acceptable flux itself is often based on critical limits for *Nmin*. Such *Nmin* thresholds have been defined as a criterion for nutrient imbalances, elevated nitrate leaching or enhanced sensitivity to frost and fungal diseases (Table 1, Spranger et al. 2004; Iost et al. 2012).

A general threshold of $Bc/Al = 1$ is a widely used criterion to derive SSMB critical loads for acidity, assuming that higher *Bc/Al* values will not damage tree roots (Spranger et al. 2004). In addition, species-specific threshold values (*Bc/Al*, Table 1) have been defined by Sverdrup and Warfvinge (1993). When the *Bc/Al* ratio falls below the thresholds, we denominate this case as exceedance of the critical limit for Al toxicity in the subsequent text.

These approaches enable the determination and mapping of critical loads for N deposition and critical loads for acidity and their exceedances for Europe (Reis et al. 2012). However, there may be a time lag between the start of exceedance of

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**Table 1** Ranges of the 'adequate to optimal' (A/O) nutritional class for tree foliar concentrations (mg g$^{-1}$) of nitrogen (*N*), potassium (*K*) and magnesium (*Mg*) as compiled by Stefan et al. (1997), ratios of *N*/*Mg* and *N*/*K* according to Mellert and Göttlein (2012); species-specific critical limits of the molar ratio of base cations to total aluminium (*Bc*/*Al*) and of the inorganic N concentration in the mineral top soil solution (*Nmin*, mg L$^{-1}$) regarding nutrient imbalances, and the critical limit for the mineral N concentration in the soil solution below the rooting zone (*Nmin*) regarding nitrogen saturation

| Species group[a] | Foliage | | | | | Soil solution | | |
|---|---|---|---|---|---|---|---|---|
| | | | | | | *mineral topsoil* | | *deepest lysimeters* |
| | *N*[a] | *K*[a] | *Mg*[a] | *N*/*Mg*[b] | *N*/*K*[b] | *Bc*/*Al*[c] | *Nmin*[d] | *Nmin*[d] |
| Spruce | 12–17 | 3.5–9 | 0.6–1.5 | 10.7–21 | 1.7–3.3 | 1.2 | 0.2 | 1 |
| Pine | 12–17 | 3.5–10 | 0.6–1.5 | 10.8–22.9 | 2–4 | 1.2 | 0.2 | 1 |
| Silver fir | 12–17 | 3.5–9 | 0.6–1.5 | 10.7–22.9* | 1.7–4* | 1.2 | 0.2 | 1 |
| Douglas fir | 12–17 | 3.5–10 | 0.6–1.5 | 10.7–22.9* | 1.7–4* | 0.3 | 0.2 | 1 |
| Other conifers | 12–17 | 3.5–10 | 0.6–1.5 | 10.7–22.9* | 1.7–4* | 1.2 | 0.2 | 1 |
| Beech | 18–25 | 5–10 | 1–1.5 | 8.2–21.8 | 1.9–3.8 | 0.6 | 0.4 | 1 |
| Birch | 18–25 | 5–10 | 1–1.5 | 8.1–21.8* | 1.7–3.8* | 0.8 | 0.4 | 1 |
| Oak | 15–25 | 5–10 | 1–2.5 | 8.1–21.8 | 1.7–3.7 | 0.6 | 0.4 | 1 |
| Other broadleaves | 15–25 | 5–10 | 1–2.5 | 8.1–21.8* | 1.7–3.8* | 0.6 | 0.4 | 1 |

[a] Forest Foliar Coordinating Centre of the Expert Panel on Foliage and Litterfall of ICP Forests (Stefan et al. 1997)

[b] Mellert and Göttlein (2012)

[c] Sverdrup and Warfvinge (1993) and Lorenz et al. (2008)

[d] Iost et al. (2012)

*Adapted

critical loads and the start of the exceedance of the underlying critical limits, as well as between exceedance of critical limits and effects, e.g. on tree nutritional status.

Foliar analyses have been used to assess the nutritional status of trees based on the concentration ranges for nutrition classes compiled from various experiments and expert knowledge by the Expert Panel on Foliage and Litterfall of the International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests) (Stefan et al. 1997) or more recently by Mellert and Göttlein (2012). Severe nutrient deficiencies may cause visual symptoms differing by species and nutrient-specific discolouration patterns (ICP Forests 2010).

The aim of this study was to carry out an exploratory investigation of the effects of high S and N deposition on tree nutrition, using recent data from intensively monitored forest plots of the ICP Forests Level II plot network (Ferretti and Fischer 2013) and currently used critical loads concepts (Spranger et al. 2004).

- Our first hypothesis was that the exceedance of critical loads for acidity and nitrogen had lasted long enough to result in an exceedance of critical limits for Al toxicity and the inorganic N concentration in soil solution.
- Our second hypothesis was that the exceedance of critical limits affects the nutritional status of trees as assessed from nutrient contents in leaves and needles.

## 2 Material and methods

The study was based on measurements of atmospheric deposition, soil solution chemistry, foliar nutrition, soil chemistry and tree growth as well as on the assessment of crown condition, carried out in the period from 2006 to 2009 on 201 forest plots of the ICP Forests intensive monitoring (Level II) network (ICP Forests 2010) as well as 43 forest plots of the similar Swedish Throughfall Monitoring Network (SWETHRO: Pihl Karlsson et al. 2011).

Bulk deposition (*BD*) and throughfall deposition (*TF*) were continuously collected in an open field and below tree canopy, respectively, at weekly, bi-weekly or in a few cases monthly sampling intervals. The bulk deposition volume was used to derive the precipitation quantity (*P*). Annual *BD*, *TF* and *P* values were calculated for each plot and year as described in Waldner et al. (2014).

Soil solution was generally sampled with suction lysimeters, typically at the same time intervals as deposition. The *Nmin* concentration and the *Bc*/*Al* ratio were calculated for each sample using the $NO_3^-$, $NH_4^+$, *Bc* and total Al concentrations. Annual mean concentrations as well as the proportion of samples ($f_{ss}$) exceeding critical limits (Table 1) were calculated for each plot, depth and year and aggregated for the top 40-cm layer of soil as specified by Iost et al. (2012). The same parameters were calculated for the depth of the deepest lysimeters per plot (from 10- to 250-cm depth).

Soil was sampled at fixed depths: 0 to 10, 10 to 20 and 20 to 40 cm, respectively, by taking about 24 samples, which were pooled to at least three composite samples per depth and analysed (dataset version AFSCDB.LII.2.1: Cools and de Vos 2010). We calculated fine earth content weighted averages for organic C/N ratio (*C/N*) and base saturation *BS* (%) between 0- and 40-cm depth as well as averages of the C/N ratios of the organic layer.

Tree density (*trees*, ha$^{-1}$) was calculated based on the growth survey typically carried out every 5 years (ICP Forests 2010).

Foliage from the upper third of the sun exposed tree canopy was sampled from at least five trees of the main tree species of each plot. In the case of deciduous species, fully developed leaves were sampled during the second half of the growing season and before the beginning of the autumn senescence. Evergreen foliage was sampled during the dormancy period. The nutritional status of the trees was assessed by comparing the mean foliage concentrations of N, magnesium (Mg) and potassium (K) to the adjacent ranges of three classes (Stefan et al. 1997) referred to as ‘low/deficient’ (L/D), ‘adequate to optimal’ (A-O) and ‘high to surplus’ (H/S) (c.f. Table 1). The foliar concentration ratios N/Mg and N/K were compared to classes of ratios established by Mellert and Göttlein (2012). The same abbreviations as for concentrations were used to refer to ratios within the adequate to optimal range (A-O), below the lower end (L/D) and above the upper end of the adequate range (H/S) (Table 1).

Crown condition and damage cause assessments of trees on the plot (typically adjacent to the foliage sample trees) include the description of the damage symptoms and the determination of the possible cause. The presence of observed symptoms was reported to various levels of detail, e.g. only symptom and cause class or Latin name of causing agent (insect, fungus etc.). When a specific symptom was reported for at least one tree in a given country for a given year, then we assumed that it would have been reported for any other tree that showed this symptom in that country and year. Consequently, trees with no mention of this specific symptom in that country and year were treated as trees showing no symptom. We focussed on the symptom ‘light green to yellow discolouration’ and calculated the proportion of trees per plot and year ($f_y$) that showed this symptom.

We used values of exceedance of the SSMB of critical loads for N deposition and for acidity that were calculated by Nagel et al. (2011), Waldner et al. (2007) and Marchetto et al. (2010) based on measurements on Level II plots.

As a simplified estimate for the exceedance of empirical critical loads for N deposition, we used the criterion that throughfall deposition of inorganic N (*TFN*, kg ha$^{-1}$ year$^{-1}$) exceeds 15 kg ha$^{-1}$ year$^{-1}$. The critical loads apply to total deposition, which is difficult to determine because of N uptake in the canopy. Total deposition is thus typically a factor of 1 to 2 times higher than *TFN*.

## 2.1 Evaluations and statistical analyses

Not all variables were available for all plots and all years for the period 2006 to 2009. The available annual values for this period for throughfall, bulk deposition, *P* and $f_{ss}$ were aggregated per plot. The values for $f_{ss}$, *N*, *Mg*, and *K* in foliage, and $f_y$ were aggregated per plot and tree species group. We used the tree species groups defined by Stefan et al. (1997) and assigned *Fagus sylvatica* (L.) including *Fagus moesiaca* (Malý) to the group referred to hereafter as ‘beech’, *Abies alba* (Mill.) and *Abies borisii-regis* (Mattf.) to ‘fir’, *Quercus robur* (L.), *Quercus petraea* (Liebl.) and *Quercus cerris* (L.) to ‘oak’, *Betula sp.* and *Fraxinus sp.* to ‘other broadleaves’, *Pinus sp.* to ‘pine’, *Picea abies* (Karst.) to ‘spruce’ and *Pseudotsuga menziesii* (Mirb.) to ‘other conifers’.

The relationship between deposition and soil solution was investigated based on these aggregates by comparing percentages of plots in three classes of the frequency of the exceedance of critical limits ($Bc/Al<1$ and $Nmin>1$ mg L$^{-1}$) in soil solution samples ($f_{SS}=0$ %, $f_{SS}>0$ % to 50 % and $f_{SS}>50$ %) for plots grouped according to exceedance of critical loads. The relationship between soil solution quality and tree nutritional status was investigated by comparing percentages of plots in tree nutritional classes as well as the frequencies of symptoms for plots grouped according to their frequency class of exceedances of species-specific critical limits in soil solution (Table 1).

Linear regression models were applied on means from 2006 to 2009 (function ‘lm’ in R Development Core Team 2009). Foliar N and Mg concentrations were used as response variables, while inorganic N and Mg concentrations in soil solution, topsoil base saturation and *C/N* ratio, tree density, precipitation, longitude, latitude and altitude were used as predictors. Terms that were (clearly) not relevant at a significance level of 90 % ($p>0.10$) were excluded from the full model. We compared models with and without inorganic N concentration in soil solution and N throughfall deposition. S deposition was not used.

The relationship between deposition and soil solution was investigated on 234 plots in total, SSMB critical loads for acidity and *Bc/Al* were available for 62 plots, SSMB critical loads for N deposition and *Nmin* in soil solution for 71 plots, and throughfall N deposition and *Nmin* in soil solution for 231 plots of which 109 had available values for *C/N*.

## 3 Results

Regarding acidification, the SSMB critical loads were exceeded at 11 out of 62 plots (17 %). Exceedance of the general critical limit for aluminium toxicity (*Bc/Al*<1) in at least one soil solution sample ($f_{ss}$>0 %) was reported from 5 of the 11 plots (45 %) where the SSMB critical load for acidity was exceeded and similarly from 27 of the 51 plots (53 %) where it was not exceeded (Fig. 1). However, the percentage of plots with exceedance of this critical limit in the majority of the soil solution samples ($f_{ss}$>50 %) was higher among the plots where the SSMB critical load for acidity was exceeded (36 %) than among the plots where this critical load was not exceeded (14 %, Fig. 1).

Regarding eutrophication, the SSMB critical load for N deposition was exceeded at 37 out of the 71 plots for which values were available for both, SSMB critical load for N deposition and *Nmin*. The percentage of plots with exceedance of the threshold *Nmin*=1 mg $L^{-1}$ in at least one soil solution sample from the deepest lysimeters was higher (65 %) among the 37 plots where the SSMB critical load for N deposition was exceeded than in the plots where it was not (38 %, Fig. 1). A similar result was found for the much larger sample of 231 plots, when throughfall N deposition >15 kg N $ha^{-1}$ $year^{-1}$ was used as a proxy for the exceedance of empirical critical load for N deposition (Fig. 1).

Values of *C/N* <25 in the organic layer of the soil were more common among the plots with throughfall N deposition >15 kg $ha^{-1}$ $year^{-1}$ (46 %), than among the plots with throughfall N deposition <15 kg $ha^{-1}$ $year^{-1}$ (36 %) (Fig. 1). Samples from the deepest lysimeters per plot with soil solution *Nmin*>1 mg $L^{-1}$ were reported for 88 % of the 16 plots with throughfall N deposition >15 kg $ha^{-1}$ $year^{-1}$ and organic soil layer *C/N*<25; for 65 % of the 46 plots with either throughfall N deposition >15 kg N $ha^{-1}$ $year^{-1}$ or organic layer *C/N*<25; and 43 % of the 47 plots with neither throughfall N deposition >15 kg N $ha^{-1}$ $year^{-1}$ nor *C/N*<25 (Fig. 1).

Conifers had foliage N concentration in the class L/D more often than broadleaved species. Higher N in foliage for spruce, pine, fir and oak as well as lower Mg in foliage for pine were observed on plots where the critical limits for *Nmin* in soil solution were exceeded compared to plots without exceedance of these critical limits (Table 2). In conifers, Mg concentrations in the L/D class were recorded only on plots where critical limits for *Nmin* in soil solution were exceeded. In beech, the percentage of plots with Mg in the L/D class was higher at plots with these critical limits exceeded compared to other plots. Similarly, the ratios N/Mg and N/K were more frequently in the H/S class in spruce and pine (Table 2, N/K not shown).

Distribution in foliar nutrition classes showed little difference between groups of plots built according to the exceedance of the species-specific critical limit for Al toxicity (*Bc/Al* <threshold) (Table 2). However, in spruce and fir, Mg concentrations in foliage were only in the L/D class on plots with exceedance of the species-specific critical limit for Al toxicity (*Bc/Al*<1.2, c.f. Table 1). Potassium was in the L/D class of spruce at four plots which had critical limits exceeded, but this was seen for only two other plots including all tree species (not shown). The ratios N/Mg and N/K were more often in the less favourable H/S class for spruce, pine, fir (only N/Mg) and

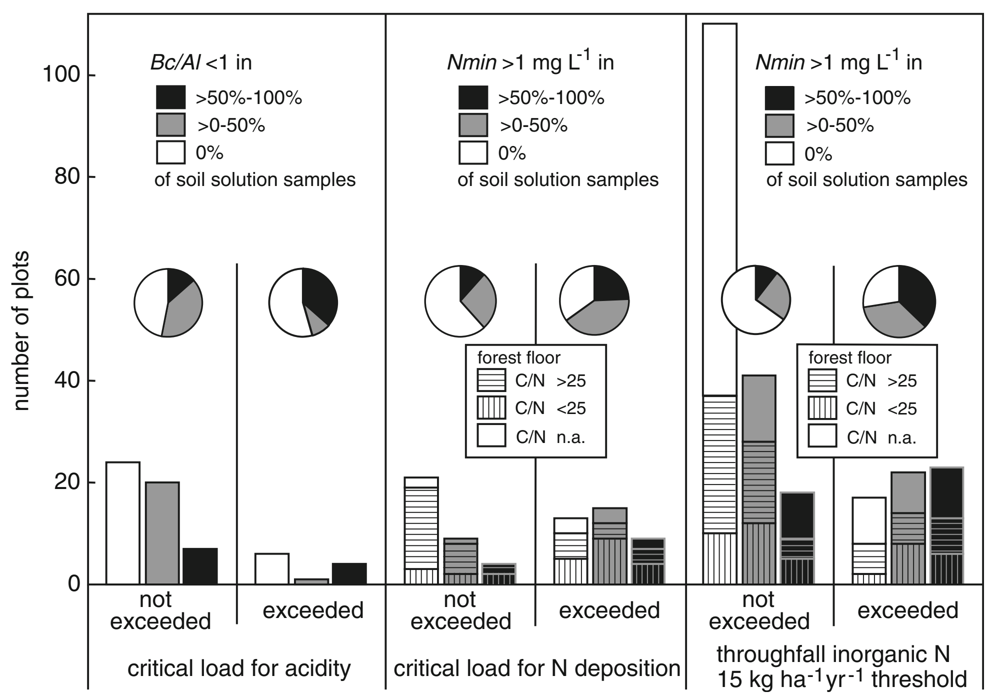


**Fig. 1** Number of plots with exceedance of the critical limits for Al toxicity (*Bc/Al*<1) and N saturation (*Nmin*>1 mg $L^{-1}$) in none ($f_{ss}$=0 %), a minority ($f_{ss}$> 0 to 50 %) and a majority ($f_{ss}$>50 %) of the soil solution samples from lysimeters in 0- to 40-cm depth and the deepest lysimeters, respectively. Number of plots (displayed as *bars*) and percentages of plots (displayed as arc length of the pie graphs disks) were compiled for groups of plots with and without exceedance of SSMB critical loads for acidity, exceedance of SSMB critical loads for N deposition and throughfall N deposition exceeding *TFN*= 15 kg $ha^{-1}$ $year^{-1}$ (*TFN*>15) based on a mean of the available annual values of the period 2006 to 2009

**Table 2** Percentages of plots in nutrition classes for groups of plots with species-specific critical limits for inorganic N (*Nmin*) and Al (*Bc/Al*<threshold) exceeded in none (0 %), in a minority (>0 %−50 %) and in the majority (>50 %) of the soil solution samples from the inorganic topsoil (0- to 40-cm depth)

| Species Group[a] | *Nmin* | *N in foliage* | | | *Mg in foliage* | | | *N/Mg in foliage* | | | Plots |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | exceedance | L/D | A-O | H/S | L/D | A-O | H/S | L/D | A-O | H/S | n |
| Spruce | 0 % | 45 | 55 | 0 | 0 | 100 | 0 | 36 | 64 | 0 | 11 |
| | >0 %−50 % | 12 | 88 | 0 | 0 | 85 | 15 | 32 | 68 | 0 | 26 |
| | >50 % | 3 | 97 | 0 | 3 | 90 | 7 | 20 | 63 | 17 | 30 |
| Pine | 0 % | 11 | 78 | 11 | 0 | 100 | 0 | 11 | 89 | 0 | 9 |
| | >0 %−50 % | 0 | 95 | 5 | 5 | 95 | 0 | 0 | 75 | 25 | 20 |
| | >50 % | 7 | 57 | 36 | 7 | 86 | 7 | 14 | 57 | 29 | 14 |
| Fir | 0 % | 100 | 0 | 0 | 0 | 100 | 0 | 100 | 0 | 0 | 1 |
| | >0 %−50 % | 14 | 86 | 0 | 14 | 43 | 43 | 86 | 0 | 14 | 7 |
| | >50 % | 0 | 100 | 0 | 0 | 67 | 33 | 100 | 0 | 0 | 3 |
| Beech | 0 % | 0 | 33 | 67 | 25 | 50 | 25 | 0 | 33 | 67 | 4 |
| | >0 %−50 % | 0 | 78 | 22 | 26 | 35 | 39 | 0 | 61 | 39 | 23 |
| | >50 % | 0 | 65 | 35 | 36 | 27 | 36 | 0 | 50 | 50 | 22 |
| Oak | 0 % | 0 | 100 | 0 | 0 | 100 | 0 | 0 | 100 | 0 | 5 |
| | >0 %−50 % | 0 | 17 | 83 | 0 | 100 | 0 | 0 | 100 | 0 | 6 |
| | >50 % | 0 | 46 | 54 | 0 | 100 | 0 | 0 | 85 | 15 | 13 |
| Species group[a] | *Bc/Al* < | *N in foliage* | | | *Mg in foliage* | | | *N/Mg in foliage* | | | Plots |
| | threshold | L/D | A-O | H/S | L/D | A-O | H/S | L/D | A-O | H/S | n |
| Spruce | 0 % | 14 | 86 | 0 | 0 | 86 | 14 | 33 | 67 | 0 | 21 |
| | >0 %−50 % | 23 | 77 | 0 | 0 | 87 | 13 | 32 | 64 | 5 | 23 |
| | >50 % | 5 | 95 | 0 | 5 | 95 | 0 | 18 | 64 | 18 | 22 |
| Pine | 0 % | 0 | 78 | 22 | 11 | 89 | 0 | 0 | 78 | 22 | 9 |
| | >0 %−50 % | 0 | 94 | 6 | 0 | 100 | 0 | 0 | 88 | 12 | 17 |
| | >50 % | 8 | 62 | 31 | 8 | 92 | 0 | 8 | 54 | 38 | 13 |
| Fir | 0 % | 29 | 71 | 0 | 0 | 71 | 29 | 100 | 0 | 0 | 7 |
| | >0 %−50 % | 0 | 100 | 0 | 0 | 0 | 100 | 100 | 0 | 0 | 1 |
| | >50 % | 0 | 100 | 0 | 50 | 0 | 50 | 50 | 0 | 50 | 2 |
| Beech | 0 % | 0 | 66 | 34 | 32 | 35 | 32 | 0 | 53 | 47 | 34 |
| | >0 %−50 % | 0 | 75 | 25 | 38 | 25 | 38 | 0 | 50 | 50 | 8 |
| | >50 % | 0 | 100 | 0 | 100 | 0 | 0 | 0 | 0 | 100 | 1 |
| Oak | 0 % | 0 | 45 | 55 | 0 | 100 | 0 | 0 | 91 | 9 | 22 |
| | >0 %−50 % | 0 | 100 | 0 | 0 | 100 | 0 | 0 | 100 | 0 | 1 |
| | >50 % | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |

Legend: *Nmin*=exceedance of species-specific critical limits for *Nmin* in soil solution; *Bc/Al*<threshold=species-specific critical limit for Al toxicity for soil solution: 0 %=exceeded in any sample, >0–50 %=exceeded in a minority of samples, >50 %=exceeded in a majority of samples. Foliar concentration ranges: L/D=low/deficient, A-O=adequate to optimum, H/S=high/surplus; n=number of plots

[a] Species grouped as suggested by Stefan et al. (1997)

beech on the plots where the species-specific critical limits for Al toxicity (*Bc/Al* below threshold) were exceeded compared with other plots (Table 2, N/K not shown).

Linear regression modelling based on the plot-wise aggregated dataset indicated that *Nmin* and *TFN* were important predictors of foliar N and Mg concentration (Table 3). For foliar N in spruce and foliar Mg in pine and fir, the explained variance increased when inorganic N concentration in soil solution (*Nmin*) and throughfall N deposition were included into the models (compare adjusted $R^2$ of $m=1$ with those of $m=2$ and 3).

Discolouration with light green to yellow foliage was more frequently reported for plots with the critical limit for *Nmin* in soil solution exceeded than for other plots, in particular for spruce (Fig. 2).

## 4 Discussion

On a European scale, this study explored and showed relations between the exceedance of critical loads and the exceedance of critical limits in soil solution, as well as tree nutritional

**Table 3** Linear regression models m to explain foliar N and Mg concentrations without (m=1) and with soil solution inorganic N in inorganic topsoil Nmin (m=2) and througfall N deposition TFN (m=3) for each tree species group

| Species group[a] | m | Estimates for intercept and regression coefficients for significant variables (at $p$<0.1) | sd | sd resid | Adj $R^2$ | n |
|---|---|---|---|---|---|---|
| N concentration in foliage | | | | | | |
| Spruce | 1 | 13.4***+0.0002 z | 1.56 | 1.56 | −0.02 | 39 |
| | 2 | 14.0***+0.614 Nmin***+0.0001 z | | 1.24 | 0.34 | |
| | 3 | 13.5***+0.520 Nmin**+0.040 TFN+0.00006 z | | 1.2 | 0.36 | |
| Pine | 1 | 11.9*−0.371 x***+0.165 y−0.008 z**+0.119 BS* | 2.41 | 1.31 | 0.65 | 27 |
| | 2 | 12.3*+0.447 Nmin −0.299 x**+0.150 y−0.006 z +0.074 BS | | 1.23 | 0.67 | |
| | 3 | 6.2+0.110 Nmin +0.203 TFN**−0.152 x+0.167 y−0.006 z*+0.077 BS | | 1.04 | 0.76 | |
| Fir | 1 | 11.5−0.0106 x+0.0635 y+0.0002 z−0.0097 BS | 0.64 | 0.27 | 0.45 | 7 |
| | 2 | 11.5−0.0040 Nmin −0.0105 x+0.0648 y+0.0002 z−0.0099 BS | | 0.27 | −0.09 | |
| Beech | 1 | 23.1***−0.137 x*+0.002 z*+0.003 trees | 1.99 | 1.63 | 0.26 | 30 |
| | 2 | 23.1***−0.022 Nmin −0.136 x*+0.002 z*+0.003 trees | | 1.63 | 0.23 | |
| | 3 | 22.9***−0.021 Nmin +0.008 TFN−0.136 x+0.002 z*+0.003 trees | | 1.63 | 0.19 | |
| Oak | 1 | 30.3***−0.00003 z−0.03148 BS−0.00729 trees | 3.49 | 2.54 | 0.29 | 13 |
| | 2 | 31.0***−0.3419 Nmin +0.0008 z−0.0388 BS−0.0087 trees | | 2.5 | 0.23 | |
| | 3 | 28.6***−0.31952 Nmin +0.14423 TFN+0.00003 z−0.02825 BS−0.00806 trees | | 2.42 | 0.18 | |
| Mg concentration in foliage | | | | | | |
| Spruce | 1 | 0.965***−0.00007 P+0.01697 CN | 0.23 | 0.21 | 0.09 | 39 |
| | 2 | 0.968***+0.00283 Nmin −0.00007 P+0.01705 CN | | 0.21 | 0.06 | |
| | 3 | 0.967***−0.0017 Nmin +0.00209 TFN−9 $10^{-5}$ P+0.01642 CN | | 0.21 | 0.04 | |
| Pines | 1 | 2.06***−0.001 P***−0.002 CN | 0.31 | 0.23 | 0.4 | 27 |
| | 2 | 2.18***+0.0671 Nmin −0.0017 P***+0.0005 CN | | 0.21 | 0.48 | |
| | 3 | 2.08***+0.095 Nmin *−0.012 TFN−0.0013 P**−0.0002 CN | | 0.2 | 0.51 | |
| Fir | 1 | 5.02*−0.0007 P−0.1768 CN | 0.57 | 0.35 | 0.41 | 7 |
| | 2 | 6.10*+0.166 Nmin −0.001 P−0.195 CN* | | 0.24 | 0.63 | |
| | 3 | 5.68*+0.132 Nmin +0.046 TFN−0.002 P−0.150 CN | | 0.16 | 0.77 | |
| Beech | 1 | 3.11*+0.1828 $Mg_{ss}$***+0.0361 x**−0.0476 y−0.0003 z+0.0034 BS | 0.51 | 0.3 | 0.6 | 30 |
| | 2 | 1.21***+0.0673 Nmin +0.1229 $Mg_{ss}$*+0.0050 BS*−0.0002 P | | 0.34 | 0.48 | |
| | 3 | 1.41***+0.0620 Nmin −0.0193 TFN+0.1368 $Mg_{ss}$*+0.0047 BS*−0.0002 P | | 0.34 | 0.48 | |
| Oak | 1 | 1.08***+0.194 $Mg_{ss}$*+0.004 BS | 0.33 | 0.23 | 0.41 | 13 |
| | 2 | 1.07***−0.008 Nmin +0.200 $Mg_{ss}$*+0.004 BS | | 0.23 | 0.34 | |
| | 3 | 1.23**−0.005 Nmin −0.010 TFN+0.194 $Mg_{ss}$+0.004 BS | | 0.23 | 0.29 | |

*m* model number, *sd* standard deviation of data (mg $g^{-1}$), *sd resid* standard deviation of residuals (mg $g^{-1}$), *adj* $R^2$ adjusted $R^2$, *n* number of plots, *$p$<0.05, **$p$<0.01, ***$p$<0.001, *x* longitude, *y* latitude, *z* lower boundary of altitude class (m), *P* precipitation (mm), *CN* C/N ratio of the inorganic topsoil from 0− to 40−cm depth, *BS* base saturation (%) in the inorganic topsoil from 0− to 40−cm depth, $Mg_{ss}$ Mg concentration in the soil solution from the inorganic topsoil from 0− to 40−cm depth, *trees* tree density (trees $ha^{-1}$), *Nmin* inorganic N concentration in soil solution (mg $L^{-1}$), *TFN* throughfall deposition of N (kg $ha^{-1}$ $year^{-1}$)

[a] Species grouped as suggested by Stefan et al. (1997)

status based on recent measurement data after several decades of high deposition loads.

In line with our first hypothesis, we observed a relationship between the exceedance of critical loads for N deposition and the frequency of elevated *Nmin* concentrations in soil solution. About half of the plots with exceedance of SSMB critical loads for N deposition showed signs of N saturation in the period 2006 to 2009. Similar results were found when we used a 15 kg $ha^{-1}$ $year^{-1}$ threshold applied to throughfall N deposition as a proxy for the empirical critical load for total N deposition on a larger number of plots. Note that total deposition is generally higher than throughfall deposition because a fraction of the deposited N is directly taken up by the canopy. Furthermore, the empirical critical load for N deposition depends on forest type and ranges from 5 to 20 kg $ha^{-1}$ $year^{-1}$. Hence, it is likely that the exceedance of the empirical critical load for N deposition is similar or even more frequent than the number of plots with throughfall N deposition >15 kg $ha^{-1}$ $year^{-1}$ may suggest. Assuming that the deepest lysimeters represent the depth of the rooting zone, it could be

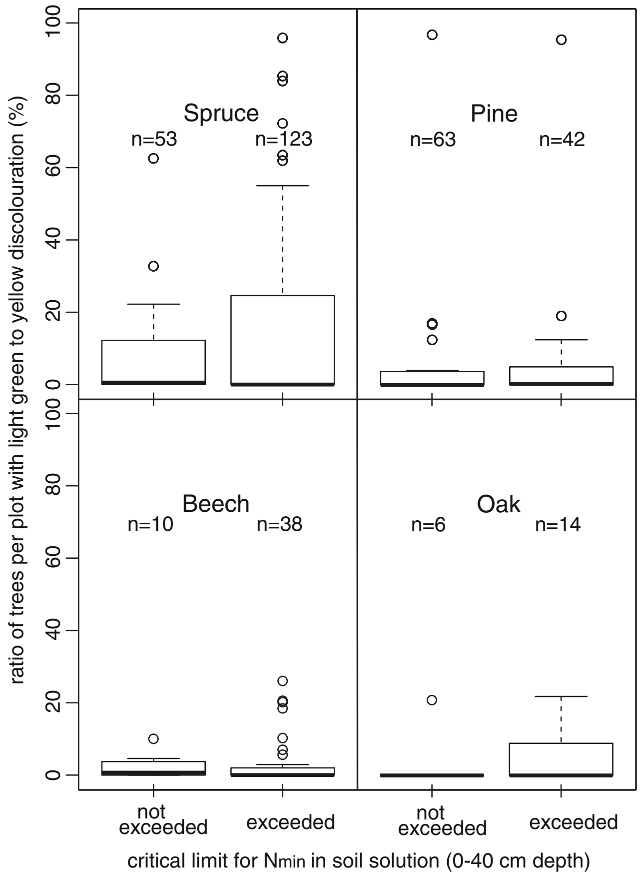


**Fig. 2** Percentage of trees per plot ($f_y$>50 %) with reported foliage discolouration (*light green to yellow*) for the species groups spruce, pine, beech and oak for plots with and without exceedance of the critical limits for inorganic N in soil solution (*Nmin*<1) in the majority of samples ($f_{ss}$>50 %) from the depth of the deepest lysimeters per plot

suggested that one third of plots with exceedance of N deposition critical loads show temporary indications and one third permanent indications of N saturation.

We found percentages of plots with insufficient or imbalanced nutrition status in the period 2006 to 2009 that were comparable to those obtained by Stefan et al. (1997) in a survey carried out in the mid 1990s. The generally lower N nutrition status of conifers compared to broadleaves may be due to the fact that conifers are more abundant at higher altitudes and latitudes, in regions with generally lower N deposition (Thimonier et al. 2010), higher rainfall and on poorer, more acidic soils.

A correlation between N deposition and N in foliage had already been found by de Vries et al. (2003) based on measurements of the period from 1994 to 1999. They reported that about 44–63, 33–71 and 26–38 % of the spatial variations in foliar N, Mg and K concentrations, respectively, were explained in a regression model that included stand age, soil type, altitude, precipitation, soil chemistry and deposition. In addition, we observed a tendency towards less favourable N/Mg and N/K ratios in foliage in high N deposition areas in the more recent data. This tendency seemed to be more strongly related to N concentration in soil solution and N deposition than to the other factors considered. The tendency towards less favourable foliage nutrition at plots with high N concentration in soil solution is in line with our second hypothesis that the exceedance of critical limits affects the nutritional status of trees. It remains to be investigated whether the temporal trends of mineral nutrition in foliage determined by Jonard et al. (2015) could be explained by changes in N deposition or N concentrations in soil solution.

The higher proportion of soil solution samples with the general critical limit for Al toxicity exceeded (*Bc/Al*<1) on plots with critical loads for acidity exceeded (Fig. 1) is in line with our first hypothesis. However, this result is based on relatively few plots with critical loads for acidity exceeded. Frequent ratios of *Bc/Al*<1 suggest acidified soils according to Sverdrup and de Vries (1994) and were found on approximately one third of these plots. However, we did not differentiate between different Al species. Determination of the Al speciation carried out for some of the plots (Graf Pannatier et al. 2011) showed that the most important toxic form, $Al^{3+}$, was typically about 30 to 100 % of total dissolved Al, whereas Hansen et al. (2007) found $Al^{3+}$ to be up to 82 to 95 % of total dissolved Al. In some cases, the $Al^{3+}$ concentration might thus be lower than total Al and less harmful than the *Bc/Al* suggests (e.g. Lange et al. 2006).

The relationship between the *Bc/Al* ratio in soil solution and foliar nutritional status was not clear, and thus, it is difficult to draw conclusions for our second hypothesis regarding acidification based on the data. The Mg and K concentrations in conifer foliage tended to be lower on plots with species-specific critical limit for Al toxicity exceeded, indicating a possible depletion of base cations due to soil acidification. No such tendency was observed for broadleaves. Values of *Bc/Al*<1 have been related to damage to fine roots but rarely related to mature forest nutritional problems, due to tree roots' ability to chelate, detoxify and prevent some of the Al from being taken up (Richter et al. 2013). Augustin et al. (2005) found stronger relationships between foliar nutrition, soil pH and base saturation than between foliar nutrition and exceedances of critical loads in an investigation of ICP Forests data from Germany, and explained this with the indirect and delayed nature of the effects. In line with this, the results of the regression analyses reported here suggested that the relationships between foliar nutrition and inorganic N concentration in soil solution and base saturation were stronger than those between foliar nutrition and throughfall N deposition.

## 5 Conclusions

This study showed that there were differences in the frequency of exceedance of critical limits for soil solution between groups of plots built according to current exceedance of critical loads, with these exceedances probably having persisted for several decades. Similar differentiation was found for tree nutritional status for groups built according to exceedances of critical limits for soil solution. The findings support the hypothesis that eutrophying or acidifying effects of inorganic N and S deposition may lead to imbalances in tree nutrition.

Further insight might be gained with supplementary analyses, e.g. by calculating SSMB critical loads for all plots, by comparing temporal trends in the variables, by using non-linear models, and by further investigation of reported symptoms.

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