

## The nutritional status of *Picea abies* (L.) Karst. across Europe, and implications for 'forest decline'

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**Summary.** During the summer of 1986, three year-classes of foliage were sampled from approximately 30-year-old Norway spruce [*Picea abies* (L.) Karst.] trees at 12 sites from S. W. Germany to N. Scotland. At sites in Germany, where trees were showing symptoms of 'decline', samples were taken from trees with 'good' crown condition and 'poor' crown condition. The distinction between 'good' and 'poor' was made on the basis of international protocols for defining crown density and foliar discoloration. There was a wide range in nutrient content (percent dry weight) in apparently healthy trees. Current year foliage had ranges of mean values per site: S(0.07–0.13%), N(0.9–1.4%), K(0.5–0.9%), Ca(0.2–0.7%), and Mg (0.05–0.1%). Ranges were greater for 2-year-old foliage: S(0.09–0.18%), N(1.0–1.8%), K(0.4–0.7%), Ca(0.2–1.4%), and Mg(0.03–0.09%). At sites with trees having 'poor' crown condition, there were significantly smaller concentrations of Mg and Ca, and larger concentrations of K in 2-year-old foliage from 'poor' trees, compared with adjacent 'good' trees. Ratios of nutrient content were more significantly related to crown condition within sites than individual nutrients, especially in older needles. 'Poor' crowns were associated with larger ratios of N:Mg, K:Mg, S:Mg, K:Ca and smaller ratios of S:K and N:K. A 'risk index' is defined for trees showing no visible 'decline' symptoms, based upon nutrient content and nutrient ratios, which may be useful in identifying sites liable to experience deterioration in crown condition.

With the exception of one German site, where few 'poor' trees were observed, the index increases from Scottish sites to English sites to Dutch sites to German sites. The index is empirical, and not necessarily related to potential effects of air pollution. The time dependence of foliar nutrient content may also be useful in diagnosis. At sites with trees having 'poor' crown condition, even apparently healthy trees showed a lack of increase in calcium content with needle age, decreases in nitrogen content and very large decreases in magnesium content with needle

age. The results show the importance of sampling several year classes of foliage from Norway spruce trees in determining the nutrient status of the tree.

**Key words:** *Picea abies* – Nutrition – Nutrient ratios – Forest decline

### Introduction

Effects on the nutritional status of trees are implicitly recognised as one of the characteristics of the 'new' type of forest decline (*neuartige Waldsterben*) affecting Norway spruce [*Picea abies* (L.) Karst.] and other tree species in central Europe. The discoloration of spruce needles, attributable to nutrient deficiencies, is used alongside non-specific needle loss in forest inventories to assess the health of forests across Europe (UNECE 1987).

Many studies have shown that the pronounced yellowing of older foliage in spruce growing on acid soils is related to magnesium deficiency (see e.g. Zöttl and Hüttl 1986; Zöttl and Mies, 1983; Zech et al. 1985; Glatzel et al. 1987). On calcareous soils, potassium deficiency may also lead to foliar yellowing (Rehfuess 1983; Tomlinson 1987). To some extent, these decline symptoms can be reversed by appropriate application of mineral fertilisers (Zöttl and Hüttl 1986, 1989), but it is not clear whether the foliar deficiency results from excessive foliar leaching, from an insufficient supply in the soil, from impaired root uptake, or from nutritional imbalances caused by pollutant input. The hypothesis originally proposed by Zöttl and Hüttl emphasised poor availability resulting from the leaching of base cations from soil and thus low base saturation of some soils. However, "predamage" and leaching of needles by exposure to photooxidants and acidic rain and fog were also suggested. Zöttl and Hüttl (1986, 1989) suggest that the increased foliar leaching of mobile elements (Mg, K, Mn and Zn) caused by exogenous factors can be compen-

sated for by increased nutrient uptake from the soil if the supply in the rooting horizons is sufficient. However, the supply of specific elements may be low in some forest soils due to degradation by natural or anthropogenic factors. On the acidic brown earths which overlie granite in the Black Forest the supply of Mg and Zn may be limited, but there is better availability of K and Mn. Similarly, limited Mg availability can occur on the different geologies of the Bavarian Forest and Fichtel Mountains, while Zn seems to have more variable availability in these two areas. In calcareous regions Mg deficiencies may be accompanied by deficiencies of Mn and on some sites, such as the moraine soils in southern Baden Württemberg, there is also evidence for K deficiency resulting from Ca:K antagonism. At some sites, displacement and leaching of base cations from the soil may lead to accumulation of toxic ions (Al, Fe and H) as well as poor nutrient availabilities, and hence to root damage and the accelerated turnover of fine roots (Ulrich et al. 1980; Krzak et al. 1988).

The role of nitrogen in influencing nutrient dynamics, and in inducing deficiencies of other nutrients such as magnesium, has been described in detail by Oren and Schulze (1989). They draw attention to the magnitude of the internal transfers of mobile nutrients within trees, regardless of the availability of elements from the soil.

Regardless of the mechanisms affecting elemental composition, concentrations of major nutrient elements in foliage give a good indication of the general health of a tree, and have been used for many years to identify the cause of visible damage to tree foliage (e.g. Binns et al. 1980; Wyttenbach et al. 1985). Furthermore the differences in elemental content which occur between year classes can be as important in diagnosing nutrient disorders as the absolute values. The transport of Mg and K from older to new foliage can determine the timing of symptom expression (Zöttl and Hüttl 1986), while differences in the accumulation of Ca, S and N in older needles can be important indications of decline symptomatology (e.g. Lange et al. 1987).

This study reports measurements of the major nutrient elements (N, K, Mg, Ca, S) for three year-classes of Norway spruce needles sampled at 12 sites during 1986 in a broad transect from S. W. Germany to N. E. Scotland. The results give an indication of the range of nutrition to be found in young forest stands (approximately 30 years old) across a wide range of soil types and climates in W. Europe. At sites in the Netherlands and West Germany, where 'decline' symptoms are apparent (needle loss and/or foliar yellowing), the relationships between visible symptoms and foliar nutrition have been investigated. The use of several age classes of needle from the same branch also gives information on nutrient distribution amongst different year classes, which may be more useful than the concentrations in a single year class for correlation with symptoms of 'decline'. The use of several year classes of needle from the same branch also minimises within-tree variability.

Wherever possible, samples were taken from existing study sites, where much background information was available, and where detailed studies could provide data over a longer period. For example, the two sites in the

Fichtel mountains in eastern Bavaria have been studied over many years, and a monograph describing the detailed measurements and interpretations drawn from these two sites has been published recently (Schulze et al. 1989a).

The use of the material from the same branch in a variety of other diagnostic tests (Cape et al. 1988, 1989; Wolfenden et al. 1988; Mehlhorn et al. 1989;) means that the results from these tests may be directly correlated with nutritional data, again minimising within-tree variability. In this respect this study is unique in the number and range of different measurements made on foliage from a single branch of the same tree, sampled at the same time. Altogether, 42 quantities were measured for most sites, not including characteristics such as mean needle length, needle loss per branch or histological examination (Cape et al. 1988).

This study is intended to complement the much more intensive research being carried out throughout Europe, in allowing comparisons to be made between sites which may differ very greatly in climate, soil type, pollutant input and expression of 'decline' symptoms. Measurements made on apparently healthy Norway spruce trees, using identical methods at all sites, give an indication of the range of foliar nutrition which can be found across Europe. Very large or very small values for nutrient content or nutrient ratios may give indications of nutritional imbalances, and a potential risk to tree health. A survey of this nature cannot be used to attribute causes for the variability between (or within) sites, as it cannot distinguish between the effects of climate, nutrient supply, genotype, air pollution or disease, but may show interesting patterns of foliar nutrition for further investigation.

## Materials and methods

*Sites.* The sites used in this study were selected for a pilot survey of potential early diagnostic indicators of forest decline, and were chosen to represent a range of pollution climates in W. Europe. Paired sites in six areas were selected, with the paired sites differing in aspect, soil type or altitude, but otherwise exposed to similar pollutant concentrations (Table 1). The site locations are shown in Fig. 1; details of exact position and descriptions of understorey vegetation and soils are given in Cape et al. (1988).

*Sampling.* At each site, between 6 and 12 trees were selected for sampling. At sites with visible damage symptoms, two strata were used ('damaged' and 'undamaged'), with equal numbers of trees sampled in each stratum. These distinctions were *relative* within a site. An attempt was made to compare the *absolute* extent of visible damage between sites by scoring each tree for leaf loss and discoloration following the UNECE protocols, which give overall damage classifications of 0, 1, 2 or 3. Trees scoring 0 or 1 are subsequently described as 'good'; those with scores of 2 or 3 are described as 'poor'. Trees on the edges of forest stands were not used. An extending 'squirrel' pruner was used to cut two branches per tree from the middle third of the canopy. This strategy is less precise than the choice of the seventh whorl, as is standard practice in W. Germany, but was necessitated by the constraints on time and personnel. The use of such pruners limited the maximum height of sampling to around 12 m; in consequence, only trees up to 15–18 m in height were included. The bias towards younger trees (<40 years) avoided the more extreme cases of visible damage, which are generally, but not always, more prevalent on older trees (>50 years), e.g. Innes and Boswell (1987).

Only green, visibly undamaged shoots were selected for analysis. Needles (10 g fresh weight) were removed from twigs in the field by

**Table 1.** The locations and pollution climates of the sites in W. Europe from which samples were taken of Norway spruce [for fuller details see Cape *et al.* (1988)]

Site code	Location	Number of trees sampled		Sampling dates 1986	Altitude (m)	Pollution climate (see key below)		
		Norway spruce				Ozone	SO <sub>2</sub> /NO <sub>2</sub>	Acidity
<i>Federal Republic of Germany:</i>								
D1	Kälbescheuer (Black Forest)	12		2–3 July	950	+++	+	++
D2	Haldenhof (Black Forest)	8		4–5 July	1000	+++	+	++
D3	S Langebranke (Harz Mountains)	8		2 Aug	600	++	++	+++
D4	N Langebranke (Harz Mountains)	8		3 Aug	600	++	++	+++
D7	Fichtelberg (E. Bavaria)	8		7 Aug	800	+++	+++	+++
D8	Selb (E. Bavaria)	8		8 Aug	600	+++	+++	+++
<i>Netherlands:</i>								
N1	Kootwijk (Central Holland)	6		7 July	50	++	++	+++*
N2	Rips (South Holland)	6		8 July	50	++	+++	+++*
<i>UK:</i>								
E1	Ashford (S. E. England)	8		2–3 Sept	50	++	++	++
E2	Winchester (S. England)	8		3–4 Sept	120	++	++	++
S1	Glenbranter (N. W. Scotland)	8		25–27 Aug	60	+	+	++
S2	Darnaway (N. E. Scotland)	8		28–29 Aug	150	+	+	+

Ozone concentrations >150 µg m<sup>-3</sup>  
 + <100 h a<sup>-1</sup>  
 ++ 100–200 h a<sup>-1</sup>  
 +++ >200 h a<sup>-1</sup>

SO<sub>2</sub>/NO<sub>2</sub> annual mean concentration  
 + <10 µg m<sup>-3</sup>  
 ++ 10–20 µg m<sup>-3</sup>  
 +++ >20 µg m<sup>-3</sup>

Acidity – wet deposition  
 + <0.25 kg H<sup>+</sup> ha<sup>-1</sup>  
 ++ 0.25–0.5 kg H<sup>+</sup> ha<sup>-1</sup>  
 +++ >0.5 kg H<sup>+</sup> ha<sup>-1</sup>

\* Rates as +++ in view of large input of NH<sub>4</sub><sup>+</sup> in rain

immersion in liquid nitrogen, then 5 g samples were washed for 10 s in chloroform to remove surface waxes and accumulated particles (Křivan *et al.* 1987). The chloroform extracts were subsequently used for the determination of amounts of epicuticular wax and surface dust (Cape *et al.* 1989).

Sampling started in S. W. Germany (sites D1 and D2) in early July 1986, and finished in England in early September 1986 (Table 1). The order of sites visited was designed to ensure the minimum differences in stage of development of new growth between sites. Current-year (1986) needles were fully expanded at all sites at the time of sampling, thereby

avoiding the large variations in nutrient content of both young and old foliage which occur prior to bud-burst and during needle expansion.

**Chemical analysis.** A 400 mg sample of the chloroform-washed needles (oven-dried at 105°C) was digested with a mixture containing H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O<sub>2</sub>/Se/Li<sub>2</sub>SO<sub>4</sub> using the method of Parkinson and Allen (1975). Details of the procedures for analysis of N, K, Ca and Mg are as given in Allen *et al.* (1974).

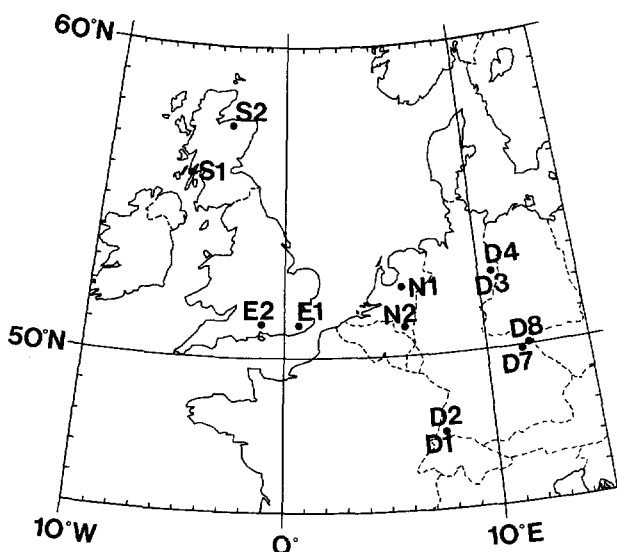
Total S was determined by X-ray fluorescence spectrometry. A pressed disc (40 mm diameter) of the powdered dried vegetation (not chloroform-washed), prepared at 20 tonnes pressure in a suitable die, was used. The spectrometer was calibrated using samples which had been analysed by wet chemical methods after digestion (Parkinson 1987).

**Statistical analysis.** Statistical differences between sites, and within sites in relation to visible symptoms of damage, were tested by analysis of variance (SAS 1985). Site differences were evaluated only for trees classified as having 'good' crown condition, taking each of the three year-classes of foliage separately, and using Duncan's Multiple Range Test for significant differences between sites. Inspection of residuals from analysis of variance indicated that untransformed variables should be used for individual elements, but that a logarithmic transform should be applied to ratios. All data for ratios were therefore log-transformed before analysis; results are presented as geometric means (i.e. back-transformed means of logarithms). Details of the factor and error terms used in analysis of variance are given in Tables 3 and 4.

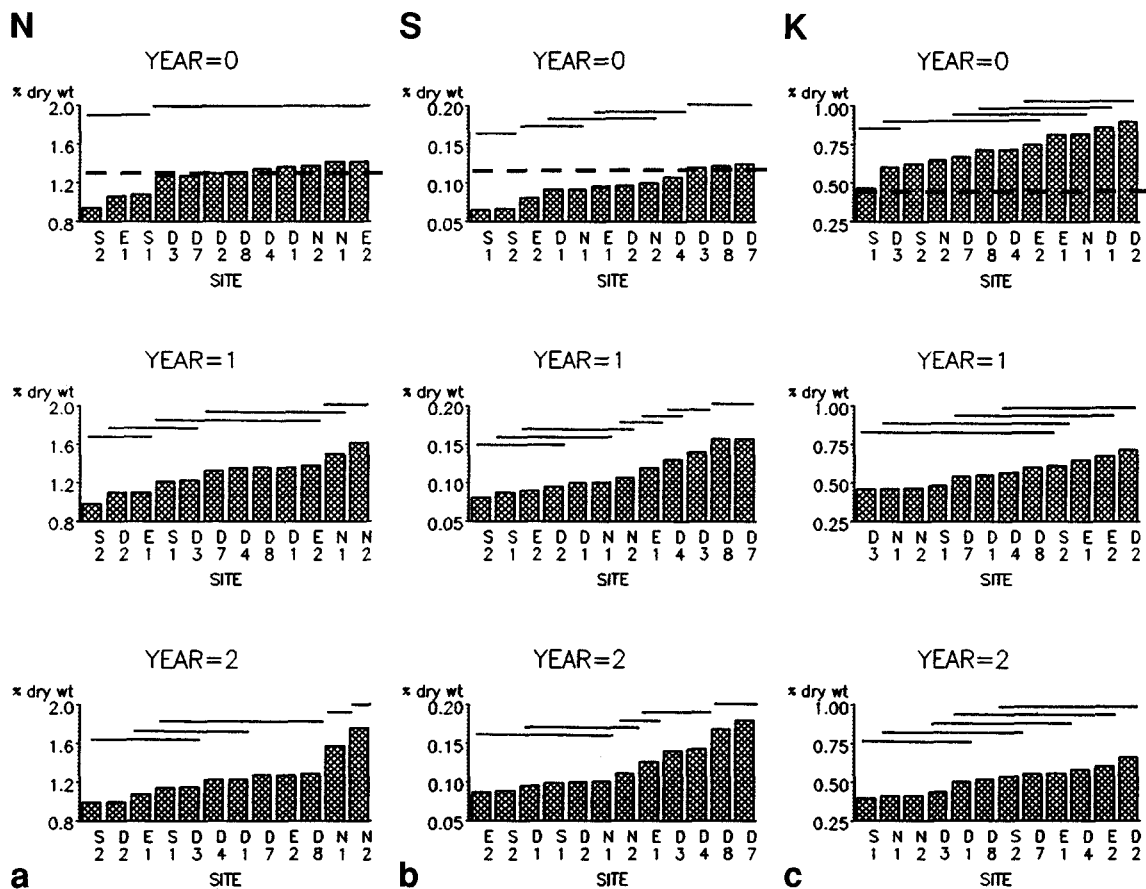
## Results

### Geographical variation

The major nutrient concentrations in each of the first three year-classes of needles are shown as site means of trees scored as 'good' (damage class 0 or 1) in Fig. 2a–e. Sig-



**Fig. 1.** Map of sampling sites used in this study (see Table 1 for site names and descriptions)



nificant differences ( $P < 0.05$ ) between sites are shown by horizontal bars. Differences are apparent in current year needles, and become more pronounced in older needles for all elements measured except potassium. For sulphur and nitrogen the concentrations within each year class range by a factor of 2 between sites, with a much larger range for calcium (a factor of 5) and magnesium (a factor of 4), but a very small range for potassium in older needles (as low as 25% for 2-year-old needles).

*Variation with needle age*

Concentrations of sulphur and calcium increase with needle age at all sites, whereas magnesium and potassium concentrations decrease with age. Nitrogen concentrations are fairly constant with age except at Dutch sites (N1 and N2) where content increases with age. The variation of nutrient content with needle age is shown in Table 2 for each site, where the change is expressed as the mean difference in percent dry weight between 2-year-old and current-year needles. It is possible that small differences in stage of development could have biased results for current year needles, but differences between 1 year-old and 2 year-old needles show a similar pattern of increases and decreases with time.

**Table 2.** Change with age in mean elemental composition of Norway spruce needles from apparently healthy trees. Values are means of the difference in % dry weight between 2-year-old and current-year needles, for  $n$  trees at each site. Site codes and descriptions are given in Table 1. Significant differences from zero are shown

	S	N	K	Ca	Mg	$n$
D1	+0.004	-0.14 <sup>a</sup>	-0.36 <sup>c</sup>	+0.097	-0.031 <sup>b</sup>	7
D2	+0.004	-0.30 <sup>b</sup>	-0.24 <sup>a</sup>	+0.024	-0.042 <sup>b</sup>	5
D3	+0.020	-0.13	-0.17 <sup>a</sup>	+0.092	-0.027 <sup>a</sup>	4
D4	+0.035 <sup>a</sup>	-0.13	-0.14	+0.056	-0.029 <sup>b</sup>	4
D7	+0.055 <sup>b</sup>	0	-0.12	+0.098	-0.021	4
D8	+0.046 <sup>b</sup>	-0.03	-0.20 <sup>b</sup>	+0.35 <sup>c</sup>	-0.012 <sup>a</sup>	7
N1	+0.010	+0.15	-0.41 <sup>c</sup>	+0.18	-0.022	4
N2	+0.011	+0.38 <sup>a</sup>	-0.24 <sup>b</sup>	+0.16 <sup>a</sup>	-0.019 <sup>c</sup>	5
E1	+0.030 <sup>c</sup>	+0.01	-0.26 <sup>c</sup>	+0.30 <sup>c</sup>	-0.002	8
E2	+0.006 <sup>a</sup>	-0.15	-0.15 <sup>a</sup>	+0.69 <sup>c</sup>	-0.029 <sup>c</sup>	8
S1	+0.034 <sup>c</sup>	+0.06	-0.08 <sup>a</sup>	+0.32 <sup>c</sup>	+0.018	8
S2	+0.022 <sup>b</sup>	+0.06	-0.09 <sup>a</sup>	+0.26 <sup>b</sup>	-0.025 <sup>c</sup>	8

<sup>a</sup>  $P < 0.05$ , <sup>b</sup>  $P < 0.01$ , <sup>c</sup>  $P < 0.001$

*Variation with visible symptoms*

The above results are for 'good' trees only. Analysis of variance with separation of variance between categories of crown health ('good' and 'poor') was possible at five sites (D1, D2, D3, D4 and D7). At other sites there were too few 'poor' trees to permit a comparison. Initial analyses showed highly significant (category\*site) interactions,

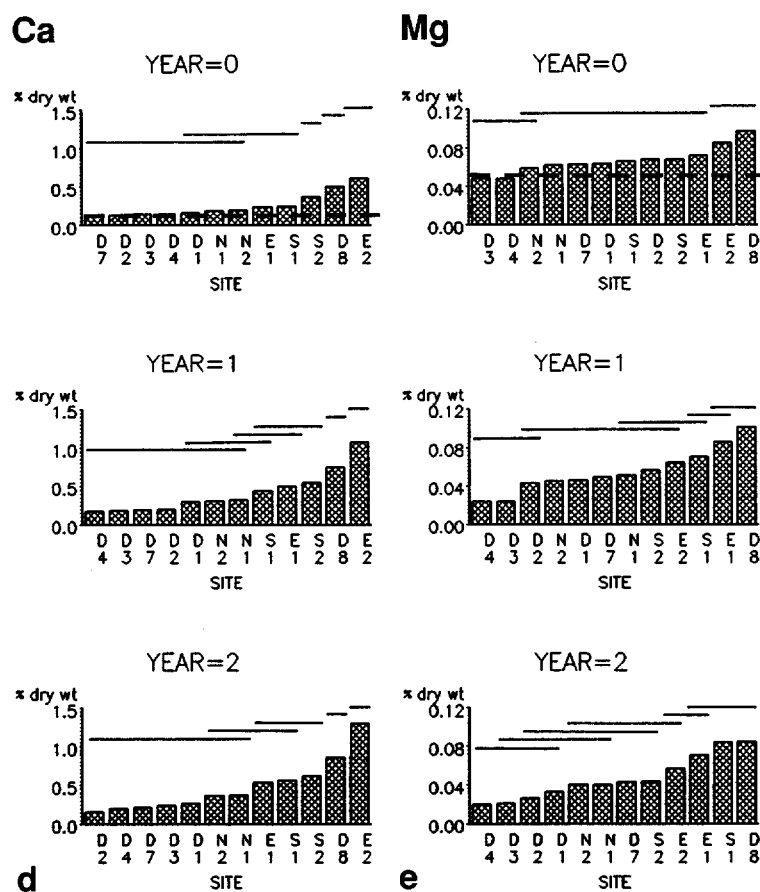


Fig. 2 a–e. Foliar concentrations (% dry weight) of nutrient elements in three year-classes of needles (0 = 1986, 1 = 1985, 2 = 1984) of Norway spruce trees showing no visible symptoms of decline. Values are means of 4 to 8 trees at each site. *Dashed lines* refer to critical levels for deficiency or excess (see Table 5). Values under the same *horizontal bar* are not significantly different ( $P < 0.05$ ; Duncan's Multiple Range Test). Site codes are shown in Fig. 1 and described in Table 1. a nitrogen, b sulphur, c potassium, d calcium, and e magnesium

and detailed inspection of the data revealed that at site D2 some of the relationships between damage classification and nutrient content were different from other sites. Nevertheless, analysis of all five sites (including D2), shows significant relationships between crown health category and nitrogen and potassium content in older needles, with larger concentrations in 'poor' trees (Table 3a).

Within individual sites there were significant differences in calcium and magnesium content, when all year classes were taken together, but this effect did not show when all sites were taken together because of the different behaviour at site D2 (Table 3b). However, exclusion of site D2 gave significantly smaller concentrations of magnesium, and larger concentrations of potassium in 'good' trees than in 'poor' trees (Table 3c).

#### Nutrient ratios

The large variation in nutrient composition between sites gives a wide range in the ratios of nutrients. The wide ranges obtained for ratios may be of greater significance than individual nutrient content, because for optimum nutrition all essential nutrients are required in appropriate proportions to nitrogen. Major differences in ratios often reflect nutritional imbalance (Ingestad and Kähr 1985). The 10 ratios derived from the five elements included in the above study are shown for 'good' trees as site means in

Fig. 3a–j. As in Fig. 2, horizontal bars denote significant differences between sites.

The ratios of nutrients are much more closely related to visible symptoms than are absolute concentrations of individual elements. Table 4 shows the results of analysis of variance by needle age, site, and crown health category. For the combined data of sites D1, D2, D3, D4 and D7, visibly damaged 'poor' trees had larger ratios of S:Mg, K:Mg, and smaller ratios of N:K in older needles than 'good' trees, reflecting the importance of magnesium, potassium and probably also nitrogen to damage symptoms.

If site D2 is excluded (as above), analysis of variance with crown health category as a factor yields a large number of significant differences (Table 4b). Some of these (including calcium) appear only in the oldest needles.

#### Discussion

##### Sampling

The sampling regime chosen for this pilot study was deliberately biased against finding differences between trees within or between sites. The objective of identifying early diagnostic indicators before the onset of visible symptoms is not well served if differences can only be found for visibly damaged foliage. Therefore, although

**Table 3 a–d.** Mean values (% dry weight) and analysis of variance for nutrient data, comparing data from trees in 'good' (*G*) and 'poor' (*P*) categories at sites D1, D2, D3, D4 and D7. Other sites had too few 'poor' trees for comparison. (a:  $P < 0.05$ , b:  $P < 0.01$ )

(a) All sites by year (factors: site, category; error: site\*category)

Year	Element	S		N		K		Ca		Mg	
	Category	G	P	G	P	G	P	G	P	G	P
0		0.11	0.11	1.32	1.32	0.77	0.79	0.14	0.13	0.059	0.053
1		0.12	0.12	1.27	1.28	0.57	0.59	0.22	0.18	0.038	0.027
2		0.13	0.13	1.17 <sup>a</sup>	1.22	0.55 <sup>a</sup>	0.61	0.22	0.18	0.029	0.021

(b) All sites, by year, excluding site D2

Year	Element	S		N		K		Ca		Mg	
	Category	G	P	G	P	G	P	G	P	G	P
0		0.11	0.11	1.33	1.32	0.74	0.79	0.14	0.12	0.057	0.049
1		0.13	0.12	1.32	1.32	0.53	0.57	0.22	0.17	0.037 <sup>a</sup>	0.024
2		0.13	0.14	1.22	1.25	0.51 <sup>b</sup>	0.59	0.23 <sup>a</sup>	0.18	0.030 <sup>a</sup>	0.020

(c) All years, by site (factors: year, category; error: year\*category)

Site	Element	S		N		K		Ca		Mg	
	Category	G	P	G	P	G	P	G	P	G	P
D1		0.095	0.091	1.32	1.29	0.64 <sup>a</sup>	0.77	0.24 <sup>a</sup>	0.18	0.047 <sup>a</sup>	0.032
D2		0.087	0.102	1.13	1.13	0.76	0.72	0.16 <sup>a</sup>	0.19	0.046 <sup>a</sup>	0.048
D3		0.133	0.121	1.22	1.23	0.50	0.50	0.19	0.17	0.031	0.030
D4		0.127	0.130	1.31	1.29	0.62	0.69	0.17	0.12	0.031 <sup>a</sup>	0.026
D7		0.154	0.158	1.29	1.38	0.59	0.62	0.17	0.15	0.051 <sup>a</sup>	0.036

(d) Years 1 and 2 only, by site

Site	Element	S		N		K		Ca		Mg	
	Category	G	P	G	P	G	P	G	P	G	P
D1		0.097	0.091	1.29	1.27	0.52	0.61	0.28	0.21	0.039	0.023
D2		0.097	0.105	1.05	1.04	0.69	0.67	0.17	0.21	0.035	0.036
D3		0.140	0.130	1.19	1.23	0.45	0.48	0.21 <sup>b</sup>	0.19	0.023	0.020
D4		0.136	0.139	1.29	1.29	0.57	0.65	0.18	0.12	0.022	0.017
D7		0.169	0.169	1.30	1.36	0.55	0.58	0.20	0.18	0.046 <sup>a</sup>	0.029

trees with visible symptoms were sampled, the foliage used for nutrient analysis was, as far as possible, green and visibly undamaged. Moreover, as visible symptoms are more likely to be seen on older trees (>50 years), the use of young trees in this study may make the detection of pre-visible symptoms more difficult.

In this study, foliage samples were taken only once during the year at each site. Mid- to late summer was chosen, to facilitate collection of samples, and to avoid times of year (e.g. immediately prior to bud-burst) when reversible discoloration of older needles is frequently observed as essential nutrients are actively transported to growing shoots (Schulze et al. 1989b). Concentrations of major nutrients tend to be most stable in late summer (e.g. Nihlgard 1987; Wytttenbach and Tobler 1988). Detailed measurements of the seasonal dynamics of foliar nutrition (Schulze et al. 1989b) suggest that for older needles,

changes are small from mid-June until October; for new needles, changes are small from July onwards. Typical variations about the mean value between July and September are: K,  $\pm 15\%$  for older needles,  $\pm 10\%$  for new needles; Ca,  $\pm 20\%$  with systematic increase; Mg,  $\pm 15\%$  in new needles, and decreasing, but  $\leq \pm 10\%$  in older needles; N,  $\pm 10\%$  in all ages of needle. While these data refer only to two sites, the scale of variability is small in comparison with the differences observed between the sites in this study: K, factor 1.6–2.0; Ca, factor 4–8; Mg, factor 2–4; N, factor 1.5–1.7. Similarly, any apparent variation in nutrient ratios caused by differences in sampling time and stage of development between sites is likely to be small in comparison with the range of ratios found. An error of  $\pm 20\%$  in each of two values would give a maximum error of  $\pm 50\%$  in the ratio. Ranges are typically factors of 6 (K/Ca), 2–6 (K/Mg) and 3–5 (Ca/Mg).

The immediate removal of needles from branches in the field, and killing of tissue by immersion in liquid nitrogen and then chloroform, prevented any possible translocation of nutrients during storage prior to analysis. Removal of the surface waxes also removed surface deposits of dust which might have interfered with analyses of Ca, Mg or K. Although needles for S analysis were not washed, the contribution of surface deposits of S to measurements of foliar S content is thought to be small (Krivan et al. 1987). Amounts of cuticular wax and surface dust removed by this process have been reported elsewhere (Cape et al. 1989). Accumulations of Ca reported here, therefore, represented the accumulations *within* the needles, rather than surface deposits.

### Nutrient deficiencies and excesses

A number of studies have defined 'deficiency' levels for major nutrients in Norway spruce (Ingestad 1959; Wyttenbach et al. 1985; Zöttl and Hüttl 1986, 1989; Krzak et al. 1988; Landolt et al. 1989). Although interactions between individual elements are important, deficiency symptoms are to be expected for element concentrations less than those listed in the third column of Table 5. Foliar yellowing, as a symptom of 'forest decline', has been associated with smaller concentrations of magnesium in needles from visibly damaged trees (e.g. Zöttl and Mies 1983). The distinction between visibly damaged and undamaged trees is at around 0.05% dry weight magnesium in 1 year-old needles. The mean magnesium concentration in 1-year-old needles was less than this at all continental sites except D8 (Fig. 2e). This is despite the bias caused by sampling apparently healthy shoots from trees showing decline symptoms. On calcareous soils potassium deficiencies can arise, but there was no evidence of potassium deficiency at any of the study sites. Indeed, as for nitrogen, there was some evidence for enhanced K concentrations in needles sampled from trees with 'poor' crown density, particularly for older needles. The data confirm the general importance of Mg, N and K content of needles to the crown condition of *Picea abies* in Europe. The tendency for larger S content at some German sites is also of considerable interest. Each of the elements is discussed in turn.

### Sulphur

Sulphur is an essential component of proteins and enzyme systems. Increases in sulphur content are associated with exposure to gaseous sulphur dioxide, (e.g. Malcom and Garforth 1977; Evers 1986; Landolt et al. 1989) and large concentrations of soil sulphate.

Although conifers can dispose of excess sulphur by re-emitting it as hydrogen sulphide (Hällgren et al. 1982), this is an energetically expensive process, and sulphur can accumulate in older needles. This is observed at all sites in the present study, but the largest absolute concentrations are observed at those sites exposed to the largest concentrations of sulphur dioxide (D7, D8) and wet deposition of sulphate (D3, D4) (see Table 1). Small concentrations are

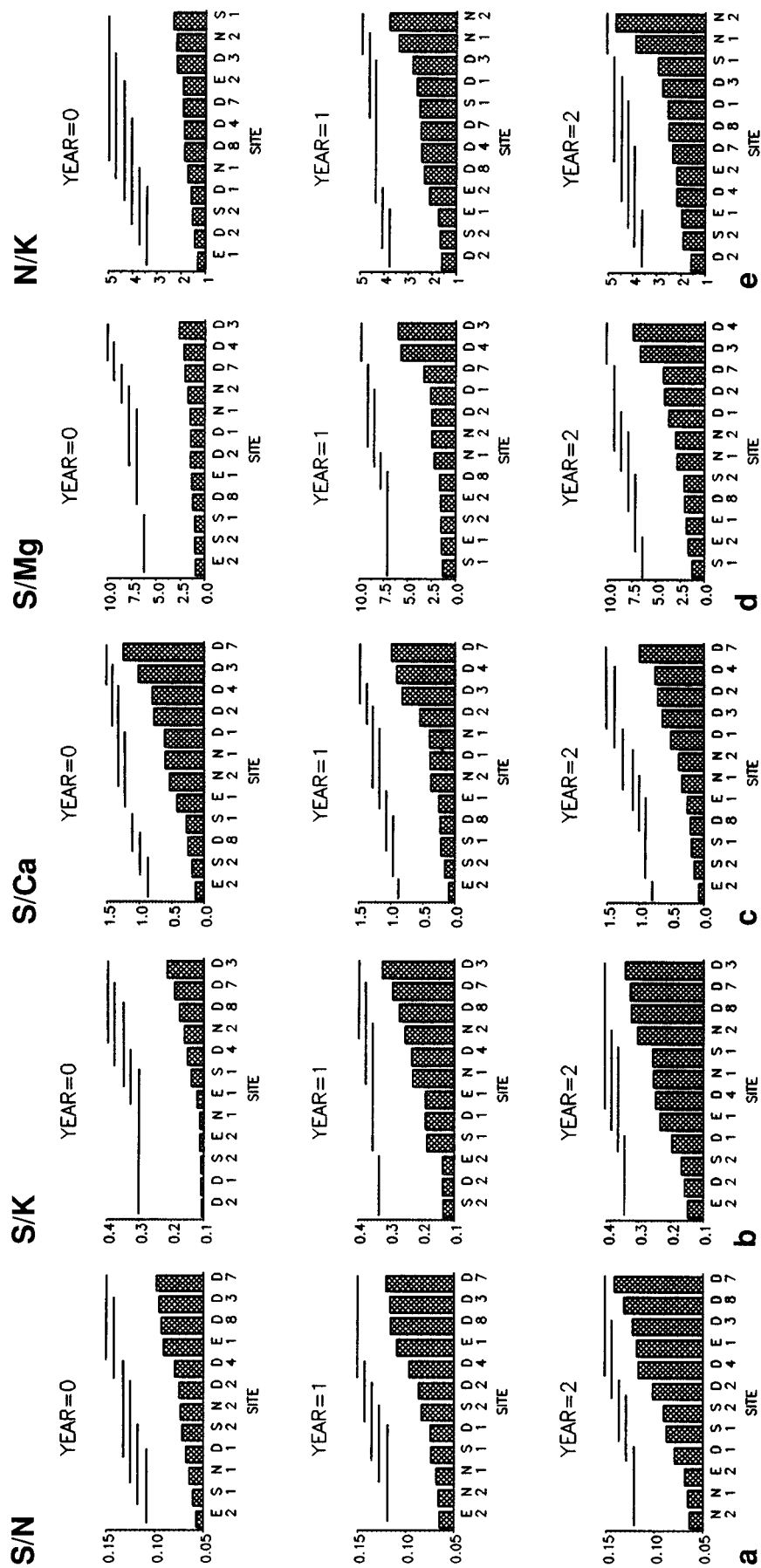
observed at the two Scottish sites and one English site (E2). The larger values in 1-year-old and 2-year-old needles at site E1 were not expected, but in the absence of reliable data on gas phase concentrations no conclusions can be drawn. Of the two Dutch sites, the more polluted (N2) shows consistently, but not significantly, larger foliar sulphur concentrations. There was no evidence of sulphur concentrations alone being correlated with decline symptoms, although sites N2, E1, D4, D3, D8 and D7 have values within the range in which pollutant inputs are considered to be having some influence on the needle nutrition of *P. abies*, between 0.12% and 0.21% (Landolt et al. 1989). There was no evidence that the rate of accumulation of sulphur was correlated with decline symptoms (Table 2).

### Nitrogen

In current-year needles, nitrogen concentrations were very similar at all sites except the two Scottish sites and the English site (E1), at which nitrogen concentrations were approaching deficiency levels (Fig. 2a). In older needles these differences were less pronounced, but the influence of widespread ammonia pollution in the Netherlands was clearly reflected in the nitrogen concentrations at both sites, with the largest increases (Table 2) at the more polluted site (N2). Excess nitrogen accumulates in needles, where it is stored as amino acids (van Dijk and Roelofs 1987). Even at sites away from extreme nitrogen inputs, decline symptoms within sites were associated with larger concentrations of nitrogen in older needles (Table 3). This observation supports the idea that nitrogen deposition in excess of forest requirements may be responsible for forest decline at some sites (Kenk and Fischer 1988; Oren and Schulze 1989). The mechanism may involve the rate of nitrogen supply relative to other essential nutrients, as discussed later (see also Ingestad and Kähr 1985). The significant decrease in nitrogen content with needle age at sites D1, D2 and E2 (Table 2) may indicate the absence of exposure to nitrogen-containing pollutants.

### Potassium

Potassium in leaves is a mobile cation, used to maintain cation balances, and important as a regulator of many physiological processes. It is also readily leached by rain from the canopy, and has the largest turnover rate of mineral nutrients. Concentrations were largest in current-year needles at all sites (Fig. 2c), and differences between sites diminished with needle age as concentrations also decreased (Table 2). There were no obvious geographical distinctions to note other than the small concentrations at Dutch sites, possibly in response to large foliar deposition of ammonium, and the large concentrations at the 'anomalous' site D2. Some of the K values approach deficiency levels (Fig. 2c) as for example, the Scottish site S1, where trees were growing in deep peat. This is perhaps not surprising since K deficiency of conifers on peats, and the use of K or P + K fertilization, is quite extensive in British forestry. The significantly larger concentrations of potassi-



**a**

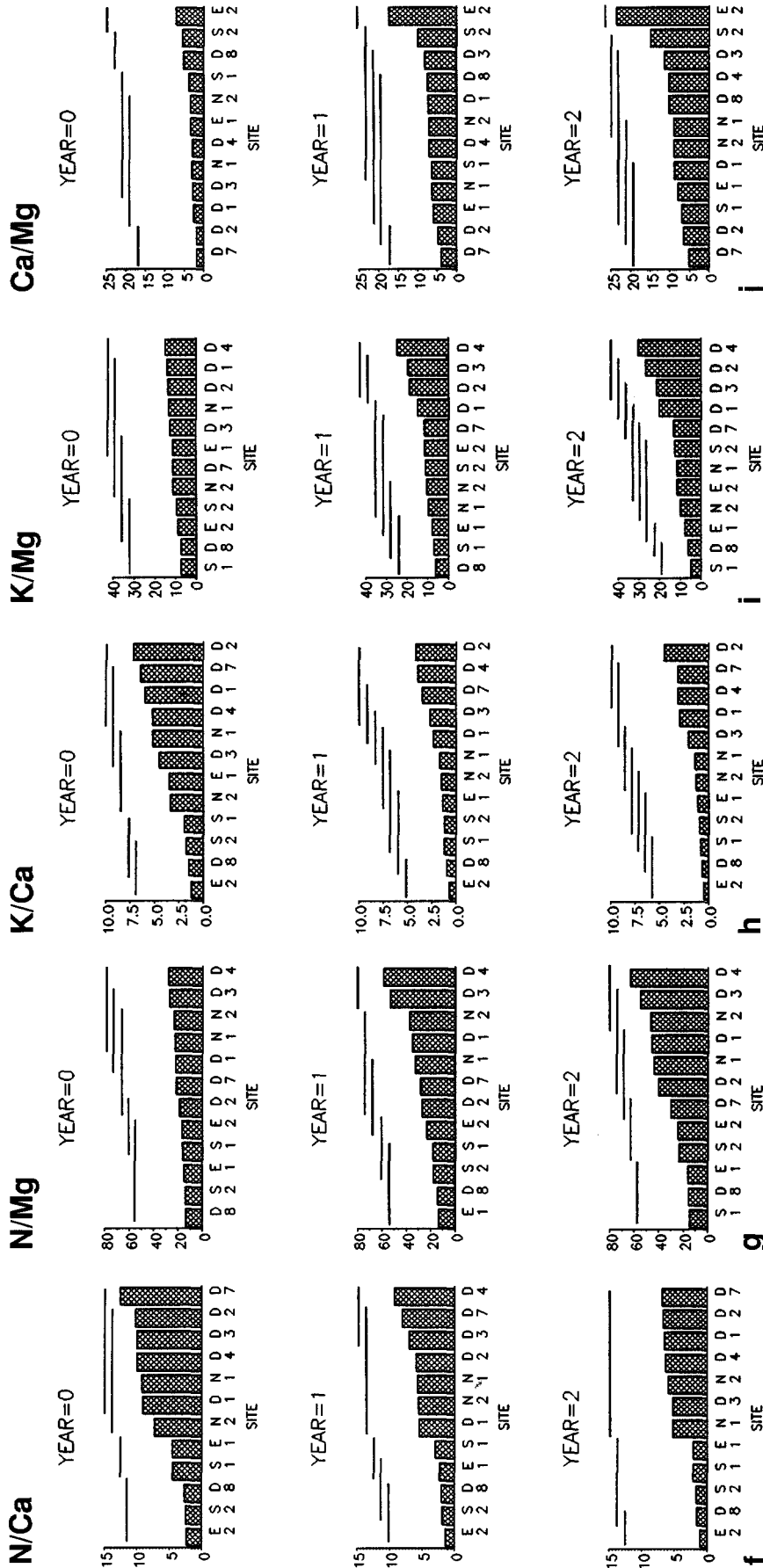
**b**

**c**

**d**

**e**





**Fig. 3.** Ratios of nutrient elements (mass/mass) in Norway spruce needles sampled from visibly healthy trees at 12 sites in W. Europe (Fig. 1) in 1986. The three year-classes refer to 1986 (0), 1985 (1) and 1984 (2). Values are means of 4 to 8 trees at each site. Values under the same horizontal bar are not significantly different. ( $P < 0.05$ ; Duncan's Multiple Range Test on log-transformed data) **a** S/N, **b** S/K, **c** S/Ca, **d** S/Mg, **e** N/K, **f** N/Ca, **g** N/Mg, **h** K/Ca, **i** K/Mg, **j** Ca/Mg. Site codes are shown in Fig. 1 and described in Table 1

**Table 4 a–d.** Geometric mean values of ratios which gave a significant difference between trees in ‘poor’ (P) and ‘good’ (G) categories in an analysis of variance for sites D1, D2, D3, D4 and D7. Other sites had too few ‘poor’ trees for comparison. (a:  $P < 0.05$ ; b:  $P < 0.01$ )

(a) All sites, by year (factors: site, category; error: site\*category)

Year	Ratio	N/K		S/Mg		K/Mg	
		Category	G	P	G	P	G
0		1.77	1.75	1.35	1.98	10.3	14.3
1		2.24	2.37	2.00	3.97	10.4	18.9
2		2.40 <sup>a</sup>	2.19	2.49 <sup>a</sup>	5.47	11.1 <sup>a</sup>	24.8

(b) All sites, excluding D2, by year

Year	Ratio	N/Mg		K/Mg		S/K		S/Mg		N/K		K/Ca	
		Category	G	P	G	P	G	P	G	P	G	P	G
0		23.7	27.5	13.0	15.8	0.147	0.144	1.91	2.27	1.83	1.74	5.28	6.46
1		39.2 <sup>a</sup>	56.6	15.5 <sup>a</sup>	23.9	0.238	0.213	3.69	5.18	2.53	2.34	2.58	3.50
2		44.9 <sup>a</sup>	64.7	18.6 <sup>a</sup>	30.3	0.253 <sup>a</sup>	0.221	4.71	6.71	2.42	2.14	2.39 <sup>a</sup>	3.55

(c) All years, by site (factors: year, category; error: year\*category)

Site	Ratio	S/K		S/Ca		N/K		N/Ca		N/Mg		K/Ca	
		Category	G	P	G	P	G	P	G	P	G	P	G
D1		0.56 <sup>c</sup>	0.121	0.574 <sup>a</sup>	0.451	2.15 <sup>a</sup>	1.75	6.25 <sup>b</sup>	8.31	30.9	45.9	2.90 <sup>b</sup>	4.74
D2		0.130	0.140	0.654	0.558	1.51	1.55	7.25	6.19	26.8 <sup>a</sup>	25.3	4.93	3.99
D3		0.276	0.247	0.760	0.723	2.52	2.50	7.34	6.95	42.1	45.6	2.75	2.93
D4		0.204	0.187	0.868	1.08	2.13 <sup>a</sup>	1.89	8.00	10.9	46.7 <sup>a</sup>	55.9	3.77	5.78
D7		0.260	0.257	0.961	1.06	2.22	2.24	8.19	8.99	25.8 <sup>a</sup>	40.2	3.70	4.18

Site	Ratio	K/Mg		S/Mg		S/N	
		Category	G	P	G	P	G
D1		14.4 <sup>a</sup>	26.2	2.24	3.17	0.072	0.069
D2		18.2	16.3	2.42	2.29	0.086	0.090
D3		16.8	18.2	4.62	4.49	0.109 <sup>*</sup>	0.099
D4		21.9 <sup>a</sup>	29.4	4.47 <sup>b</sup>	5.60	0.096	0.099
D7		11.6	17.9	3.02 <sup>a</sup>	4.60	0.117	0.114

(d) Years 1 and 2 only, by site

Site	Ratio	S/N		S/K		S/Ca		S/Mg		N/K		N/Ca	
		Category	G	P	G	P	G	P	G	P	G	P	G
D1		0.075 <sup>a</sup>	0.070	0.188 <sup>b</sup>	0.147	0.397	0.482	2.78 <sup>a</sup>	4.23	2.50 <sup>b</sup>	2.10	5.28 <sup>a</sup>	6.90
D2		0.093	0.100	0.143	0.154	0.605	0.530	3.13	3.01	1.54	1.55	6.15	5.33
D3		0.118	0.106	0.321 <sup>a</sup>	0.276	0.697	0.726	6.24	6.52	2.73	2.59	5.92	6.84
D4		0.105	0.106	0.238	0.210	0.774	1.13	6.37	8.31	2.26	1.98	7.35	10.7
D7		0.129	0.123	0.308	0.290	0.890	0.942	3.70 <sup>a</sup>	6.07	2.39	2.35	6.90	7.49

Site	Ratio	N/Mg		K/Ca		K/Mg		Ca/Mg	
		Category	G	P	G	P	G	P	G
D1		37.0 <sup>a</sup>	60.3	2.11 <sup>a</sup>	3.29	14.8 <sup>a</sup>	28.8	7.01 <sup>a</sup>	8.77
D2		31.8	30.2	4.13	3.44	21.3	19.5	5.18	5.67
D3		53.1	61.4	2.17	2.64	19.4	23.7	8.96	8.98
D4		60.4	76.2	3.25	5.40	26.7	38.2	8.22	6.73
D7		28.7 <sup>b</sup>	49.2	2.89	3.30	12.0	20.9	4.16 <sup>a</sup>	6.26

**Table 5.** The foliar concentrations, expressed as percentage of total needle dry weight, of N, K, Ca, Mg and S for current year *Picea abies* shoots at which visible deficiency symptoms occur and the deficiency, the intermediate or normal, and the optimum ranges for growth. Based on Zöttl and Mies (1983), Burg (1985) and Landolt et al. (1989)

Element	Foliar concentration (%)			
	Visual deficiency symptoms	Deficiency range for growth	Intermediate or normal	Optimum
N	0.8 →1.3	1.0 →1.6	1.08→2.4	1.7 →2.4
K	0.14→0.4	0.2 →0.7	0.4 →1.21	0.7 →1.23
Ca	–	0.13→0.30	0.10→1.26	0.3 →0.68
Mg	0.02→0.05	0.03→0.07	0.05→0.31	0.07→0.14
S	–	–	0.03	0.12→0.21

um in older needles of 'poor' trees is in contrast to the normal decrease of K content with needle age as shown by the values for 'good' trees. Larger potassium concentrations may reflect deficiencies in other cations such as magnesium, with potassium serving to maintaining the charge balance.

### Calcium

Calcium is largely immobilised in cell walls, and would be expected to accumulate with time, although there is some evidence that calcium may be partly mobile in Norway spruce (Schulze et al. 1989b). In areas exposed to SO<sub>2</sub>, crystals of gypsum (CaSO<sub>4</sub>) have been observed on leaf surfaces presumably as the result of reaction between leached calcium ions and SO<sub>2</sub> (Rehfuess 1983). Such deposits would not have been recorded using our methods, which removed surface material before analysis, and there is no evidence that internal calcium concentrations were related to exposure to SO<sub>2</sub> or sulphate pollutants. Concentrations increased with time at most sites (Fig. 2d, Table 2). The very large accumulations at sites D8 and E2 reflect supply of the element; at site D8 (Selb) the trees were growing in soil relatively richer in available calcium than at site D7 (Oren et al. 1988), while trees at site E2 (near Winchester) were growing on chalk bedrock. The relative rate of accumulation, however, was no greater at these sites than at others where calcium supply was adequate (Table 2). The very small concentrations at other German sites were close to deficiency levels (Fig. 2d, see also Krzak et al. 1988), and increases with age were small or non-significant. The rather slow rate of accumulation of calcium in leaf tissue may be a useful marker of deficiency, as noted by Schulze et al. (1989b). The comparison of different age classes of needle from the same branch removes much of the internal variability and may be a more reliable measure than absolute concentrations. At two sites (D2 and D3), concentrations of calcium were significantly greater in 'good' trees than in 'poor' trees, indicating that the symptoms were related to Ca supply.

### Magnesium

Magnesium is associated with carbon assimilation, being the essential metal in chlorophyll, and necessary for the correct functioning of the phloem transport system in leaves. The ion is mobile in plant tissue, with older needles acting as storage reservoirs from which the ion can be translocated to new growth. Magnesium can also be leached from leaves by rain, although the rate of turnover (leaching as a proportion of total tree content) is much less than for potassium. Initially large and fairly uniform concentrations among sites diminished rapidly with age, except at sites in Britain and site D8 (Fig. 2e). The strong maritime influence on rain chemistry in Britain is responsible for the deposition of considerable quantities of magnesium derived from sea-salts which may offset any tendencies to deficiency in soils (Roberts et al. 1989). At site S1, close to the west coast of Scotland, foliar magnesium concentrations increased significantly with needle age (Table 2), possibly reflecting the large inputs of marine-derived salts in this region (Cape et al. 1984). As noted above, the site D8 was relatively rich in available magnesium and calcium. By contrast, older needles even from visibly 'good' trees at other German and Dutch sites approach deficiency levels. With the exception of site D2, 'good' trees had significantly larger concentrations of magnesium than did 'poor' trees, so that the observation of needle discoloration attributable to magnesium deficiency is not surprising at these sites.

Although magnesium deficiency in trees has been clearly demonstrated as the likely cause of the observed yellowing symptoms in exposed needles, through 'diagnostic fertilization' trials (Zöttl and Hüttl 1989), it is not yet clear whether deficiencies are primarily caused by deficiencies of supply in the soil, by inhibition of root uptake by aluminium ions in acidified soil (Ulrich et al. 1980), or induced by excessive supply of nitrogen (Oren and Schulze 1989). There is, however, increasing evidence that extremes of climate, pollutant inputs and soil conditions all may contribute to magnesium deficiency of foliage, dependent on site, region and tree provenance (Roberts et al. 1989).

### Element ratios

*Ratios involving sulphur.* The ratio of sulphur to nitrogen has been suggested as a potential indicator of pollutant exposure to SO<sub>2</sub> (Malcolm and Garforth 1977). On a continental scale, with the range of pollution climates covered in this study, this ratio is perhaps better used as an indicator of the relative importance of S and N compounds as components of atmospheric pollution. The smallest values (Fig. 3a), at Dutch sites, are a consequence of the very large nitrogen inputs. By contrast, sites D3, D4, D7 and D8 give large ratios which appear to reflect large atmospheric inputs of S rather than differences in soil type, or the availability of other nutrients. Ratios of S/N at site D8, for example, were similar to those at D3, D4 and D7 in spite of much greater Ca and Mg concentrations at that site (Fig. 2d, e). There was, however, no evidence of a relationship between S/N ratios and visible decline symptoms; this

ratio appears to be a characteristic of the site rather than an indication of tree health.

The ratio of sulphur to potassium increases greatly with needle age, as S increases and K decreases (Fig. 3b). The geographical pattern again probably reflects supply of sulphur, with the more polluted Dutch site (N2), and the German sites (D8, D7 and D3) at which atmospheric sulphur inputs are thought to be important, showing the larger values. The large value for site S1 reflects rather small K concentrations, whereas the other British sites and both sites in the Black Forest (D1 and D2) have small S/K ratios, corresponding to the smaller inputs of S as SO<sub>2</sub> or sulphate. As for S/N, the ratio S/K appears to reflect site factors rather than tree health, except where decline symptoms are strongly associated with increased concentrations of potassium in older needles (D1 and D3).

The ratio of sulphur to calcium is more stable with time, as concentrations of both elements tend to increase with needle age (Fig. 3c). The lowest values at E2 are caused by large calcium concentrations. In general, values of the ratio are smallest at British sites and largest at German sites, with Dutch sites intermediate. The relatively low values at site D8 are also associated with large Ca concentrations. In older needles, larger S/Ca ratios were associated with decline symptoms at sites where both 'good' and 'poor' trees were sampled (Table 4). The differences across sites of a factor of over 10 suggest that this ratio may be useful for determining sites 'at risk'. Moreover, there is a potential mechanism for the creation of large S/Ca ratios if sulphate deposition leads to acidification and enhanced leaching of calcium from forest soils.

The ratio of sulphur to magnesium, as for S/K, increases rapidly with needle age (Fig. 3d). Although differences across sites vary by only a factor of 5, there are highly significant relationships between S/Mg ratios and tree health in 1- and 2-year-old needles (Table 4). One can speculate that this ratio also reflects the underlying nutrient stress caused by sulphate leaching of magnesium from soils.

*Ratios involving nitrogen.* The ratio of nitrogen to potassium (Fig. 3e) has been associated with sensitivity to frost (Puempel et al. 1975), with larger values inducing greater sensitivity. Larger values of this ratio within sites were associated with healthy trees, particularly in older needles (Table 4). Nitrogen accumulation relative to potassium was also seen for Dutch sites. The large values for site S1 are caused by low potassium concentrations, as noted earlier. The observed higher values for this ratio in healthy trees suggest that predisposition to winter injury is not the cause of the decline symptoms at these sites.

In contrast, the ratios of nitrogen to calcium and magnesium are significantly greater for 'poor' trees (Table 4), reflecting the underlying influence of calcium and magnesium nutrition (Table 3). Nitrogen: calcium ratios decrease with needle age, as calcium accumulates and nitrogen concentrations fall (Fig. 3f). There is, however, a clear distinction in older needles between continental sites which gave values for the ratio greater than 5, even in apparently healthy trees, and British sites (and D8) where the ratio was 2 or less.

A similar pattern is observed for nitrogen:magnesium ratios, but here the ratio increases with time at all sites as magnesium concentrations fall (Fig. 3g). The distinction between sites with decline symptoms and those without is less marked than for N:Ca ratios, even though there is still a significant difference between 'poor' and 'good' trees within sites (Table 4). Relatively larger values for N:Mg and N:Ca ratios have been noted along a gradient of ammonia/ammonium pollution in Sweden (Nihlgard 1987) with larger values at more exposed sites. An imbalance of nitrogen and magnesium has been suggested as the cause of the development of foliar yellowing, where N:Mg ratios in current-year needles at a declining site were consistently above a mass ratio of 21, below which optimum growth of spruce seedlings can be expected (Schulze et al. 1989b). This ratio was exceeded at all continental sites except D8 (Fig. 3g).

*Ratios of 'base cations' (K, Ca, Mg).* Larger values of the ratios of potassium to calcium and potassium to magnesium were both associated with decline symptoms within sites (Table 4). In apparently healthy trees at sites where decline symptoms were evident these ratios were also larger than at other sites (Fig. 3h, i). As discussed earlier, this relative increase in potassium content, particularly in older needles, may reflect the role of potassium in maintaining a charge balance in trees deficient in magnesium and calcium. As such, the ratio is likely to be a more sensitive indicator of nutrient stress than concentrations of single nutrient elements. There was no correlation of calcium:magnesium ratios with decline symptoms, and the ratio was comparatively constant across sites with the exception of site E2 where trees were growing on chalk (Fig. 3j).

## Conclusions

At the start of this study it was not clear whether within-site variability would be so great that no significant differences would be found between sites, given the relatively small numbers of samples that were experimentally practicable. However, clear differences between sites have been shown for apparently healthy trees, illustrating the range of nutrient composition of Norway spruce needles across a variety of soils, climates and genotypes in W. Europe. Many factors may be responsible for differences between sites, including atmospheric inputs: magnesium in rain in Scotland, large sulphur concentrations (in air, rain and/or soil) in the east of W. Germany, and large nitrogen concentrations (both gaseous ammonia and ammonium in rain) in the Netherlands. Soils are also important, notably the influence of chalk bedrock on calcium concentrations in south-central England, the lack of potassium in deep peat soils of W. Scotland and the general better availability of magnesium and calcium at one site (D8) compared to one nearby (D7). The lack of magnesium and calcium in upland forests of W. Germany may also be soil-related, as is clear from the continuing debate over the primary factors responsible for declines of Norway spruce. The effect of differences in

sampling date between sites is thought to be small relative to other factors.

Comparisons within sites of trees with 'good' or 'poor' crown condition showed that 'poor' crowns were associated with smaller concentrations of magnesium and calcium and larger concentrations of potassium, but it was not possible to identify 'critical' concentrations above or below which crown conditions deteriorated at all sites. The content of individual nutrients is not uniquely associated with crown condition across the range of sites studied, although there were significant differences within sites in nutrient content related to crown condition.

Although for individual nutrients significant differences between sites were detected, ratios of nutrient content covered a much wider range, up to a factor of 10 for sulphur: calcium ratios in apparently healthy trees. Within sites significant differences between 'good' and 'poor' trees were observed, with 'poor' trees having larger ratios of N/Mg, K/Mg, S/Mg and K/Ca, and smaller ratios of S/K and N/K. As for individual nutrients there were no clear-cut critical values associated with 'poor' crowns.

One of the objectives of this study was to identify pre-visible symptoms which could be useful in diagnosing risk. If it is assumed that apparently healthy trees growing at sites where damage symptoms are present are also under stress, then nutrient data from these apparently healthy trees may be useful in establishing criteria for risk of damage. The sites exhibiting significant damage symptoms (D1, D2, D3, D4, D7) were often a distinct group in the range of variation of nutrient content and nutrient ratios (see Figs. 2, 3).

Approximate risk thresholds may therefore be defined for older needles (1- or 2-years old), with a higher risk associated with: (1) calcium below 0.3%, (2) magnesium below 0.05%, (3) sulphur: calcium above 0.4, (4) sulphur: magnesium above 3, (5) nitrogen: calcium above 5, (6) nitrogen: magnesium above 30, (7) potassium: calcium above 2, and (8) potassium: magnesium above 15.

As an illustration of this method of risk assessment, using these eight threshold values, one may construct a simple risk index (R. I.) for each site, defined as:

$$R. I. = \frac{1}{8n} \sum_{i=1}^n (\text{number of thresholds exceeded})_i$$

where  $n$  is the number of trees randomly sampled at a site and  $i$  denotes the  $i^{\text{th}}$  tree.

R. I. is then a number between 0 and 1, where 0 denotes no risk thresholds exceeded, and 1 denotes all 8 risk thresholds exceeded on every tree. The results of this calculation for 1-year-old needles from apparently healthy trees in this study are shown in Table 6. It should be noted that although the risk index (R. I.) as defined above is not directly related to air pollution effects, half of the risk thresholds are associated with larger concentrations of sulphur or nitrogen in foliage, and all involve small concentrations of calcium and magnesium in foliage. The risk index does not give any indication of when or if visible symptoms will occur, but may identify areas worthy of more detailed investigation and monitoring.

**Table 6.** Risk of occurrence of damage symptoms, based upon threshold values of nutrient composition and nutrient ratios in 1-year-old Norway spruce needles (see text). The risk index (RI) is the mean fraction of threshold values exceeded per tree. Values of R. I. are derived from apparently healthy trees at each site. Site codes and descriptions are given in Table 1. A value of R. I. of 0 denotes low risk, and a value of 1 denotes high risk

Site	R. I.	No. of trees
D1	0.48	7
D2	0.69	4
D3	0.94	4
D4	0.97	4
D7	0.69	4
D8	0.02	7
N1	0.38	4
N2	0.45	5
E1	0.02	8
E2	0.11	8
S1	0.02	8
S2	0.03	8

Finally, additional information may be obtained by comparison of nutrient content of different year classes of needle. Apparently healthy trees at 'high risk' sites showed decreases in nitrogen content, a lack of significant increase in calcium content, and large decreases in magnesium content in a comparison of current-year and 2-year-old needles (Table 2). When data for all nutrients are not available (e.g. sulphur), the nutrient content of different age classes of needle may give an indication of sites 'at risk'.

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