

Chapter 16

Beech foliar chemical composition: A bioindicator of air pollution stress

G. Amores, J.M. Santamaría*

*Department of Chemistry and Soil Science, University of Navarre, Iruñlarrea s/n,
31080 Iruña-Pamplona, Spain*

Abstract

During the summers of 1995 and 1997, 238 foliar samples were taken from beech (*Fagus sylvatica* L.) trees in 17 stands belonging to the two most representative vegetation series found in Navarre (western Pyrenees, Spain). Each unwashed sample was analysed for calcium (Ca), copper (Cu), iron (Fe), potassium (K), magnesium (Mg), manganese (Mn), nitrogen (N), sodium (Na), phosphorus (P), sulphur (S), and zinc (Zn). Defoliation of sampled trees was also assessed.

Data analysis showed Ca, Mg, and S concentrations exceed the values reported in literature, while Fe and Cu concentrations were below such references. Main macronutrient ratios also exceeded referenced values, a possible cause of nutritional imbalances in the sampled trees. A low concentration of microelements of anthropogenic origin shows that atmospheric pollution caused by such pollutants is very low in this area, although a decreasing gradient from NW to SE can be observed, probably due to long-range transport from emitter points situated in the Bay of Biscay area.

1. Introduction

The decline of forests induced by air pollutants, global climate change, and their interaction with traditional diseases and pathogens over the last few decades, clearly indicates the need for urgent measures to protect our forests.

In this context, an evaluation of the nutritional status of trees and quantification of pollutants can be one of the most powerful diagnostic tools to determine their condition (Innes, 1993; Bussotti et al., 1995; Anonymous, 1997).

Nevertheless, although several authors have measured element concentrations in beech (*Fagus sylvatica* L.) leaves (Brumme et al., 1992; Fischer et al., 1993; Szarek et al., 1993; DeVries et al., 1995; Maňkovská, 1997, 1998;

*Corresponding author.

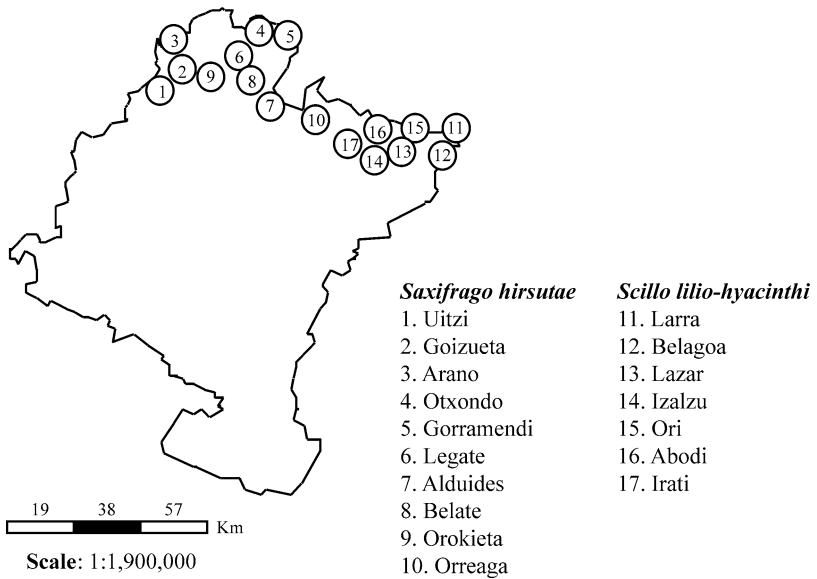


Figure 1. Geographic location of selected beech stands in Navarre.

Heinze, 1998; Meiwes et al., 1998), their conclusions when relating their results to the influence of anthropogenic pollution on forest health are varied and even contradictory.

Beech is the most important deciduous forest species in Navarre, covering an area of 136 291 ha, or 39% of the forest cover. Therefore, in this study, our aim was to assess the condition of Navarre's beech forests and establish its possible relationship to foliar nutrient concentrations and ratios, studying their validity as potential indicators of forest health.

2. Materials and methods

Seventeen forest stands, distributed throughout Navarre (western Pyrenees, Spain), were selected. These forests belong to the two most representative vegetation series found in this region (Rivas-Martínez et al., 1991), namely, *Saxifraga hirsutae*-*Fageto sylvaticae* S. (acidophilic) and *Scillo lilio-hyacinthi*-*Fageto sylvaticae* S. (basophilic). Forests belonging to the first vegetation series developed on silicon (Si)-rich oligotrophic soils, and those belonging to the second series developed on Ca-rich soils (Fig. 1).

Sampling was carried out over the summers of 1995 and 1997, in late August and early September. In all stands, mean amount of precipitation and mean air temperature were higher during the 10 months preceding the second sampling

(1997) than during those preceding the first sampling (1995). For precipitation, the differences in June, July, and August were particularly pronounced (Anonymous, 1994–1997).

Seven trees of the same canopy class (dominant or codominant trees according to Kraft classification) were chosen in each forest stand. Foliage samples were taken from the upper third of the crown (wind- and light-exposed), using a telescoping tree pruner. The unwashed samples were dried at 60 °C and then ground in a mill.

To determine the concentrations of Ca, Cu, Fe, K, Mg, Mn, Na, P, S, and Zn, the dried samples were digested with a mixture of concentrated HNO₃ and HClO₄ (2:1), via wet digestion. A small quantity of concentrated HF was also added, in order to destroy the silica. All chemicals used were of Suprapur Grade. Digestions were gauged with Milli-Q quality water (resistivity < 16 MΩ cm). Ca, Cu, Fe, Mg, Mn, and Zn concentrations were determined by means of atomic absorption spectrophotometry, and K and Na concentrations by atomic emission spectrophotometry, using a Perkin Elmer AAnalyst 800 spectrophotometer. The concentrations of P and S were determined by ICP (Jovin Ibon J-38S). Nitrogen concentration in foliage samples was determined by the Kjeldahl method. The performance of all methods was verified by analyzing certified reference material (CRM-*Fagus sylvatica*) and no method bias was detected.

Crown condition assessment of the selected trees was carried out according to the rules detailed in the UN/ECE (1997) Manual.

Relationships between element concentrations, ratios, and site-specific data were examined with Pearson's *r* test (Bonferroni's adjustment) and Factor Analysis. The significance of differences between groups of data was assessed by one-way analysis of variance. All calculations were done in SPSS, v. 9.

3. Results

Results corresponding to both sampling years were pooled in the same group for each stand, as conclusive differences were not obtained by statistical analysis of the data.

In general, crown condition of Navarre's beech forests is quite good, and only three of the stands (Otsondo, Orreaga, and Larra) are moderately defoliated (defoliation > 25%). The rest of the stands are only slightly defoliated (defoliation = 11–25%).

Concerning element concentrations in foliage, we compared our results to various data cited in the literature for beech (Maňkovská, 1998; Flückiger and Braun, 1999) (Table 1).

For macronutrient concentrations, many of the values exceed the maximums shown in the cited literature. Calcium concentrations were higher in all stands,

Table 1. Yearly rainfall (mm), defoliation (%), and foliage element concentrations (mean \pm S.D.) in beech stands (* literature data reported for beech)

Stand	Rainfall	Defoliation	Macronutrients (mg/g)						Microelements (μ g/g)				
			Ca	K	Mg	N	P	S	Cu	Fe	Mn	Na	Zn
Uitzzi	1852	12.7 \pm 6.3	9.7 \pm 1.4	6.0 \pm 1.7	1.8 \pm 0.4	22.9 \pm 2.6	1.3 \pm 0.3	2.1 \pm 0.3	7.0 \pm 1.0	107 \pm 34	453 \pm 199	232 \pm 89	35 \pm 11
Goizueta	2032	18.1 \pm 5.6	10.1 \pm 2.4	8.5 \pm 2.3	1.1 \pm 0.5	21.4 \pm 3.6	1.4 \pm 0.4	2.0 \pm 0.3	6.6 \pm 1.7	177 \pm 61	419 \pm 131	517 \pm 250	38 \pm 12
Arano	2179	17.5 \pm 3.3	11.5 \pm 4.7	7.0 \pm 1.6	1.7 \pm 0.5	21.2 \pm 2.1	1.5 \pm 0.3	2.1 \pm 0.3	6.3 \pm 1.0	91 \pm 63	409 \pm 202	1097 \pm 413	48 \pm 15
Otsondo	1991	26.1 \pm 5.6	12.2 \pm 3.6	6.9 \pm 2.2	2.1 \pm 0.4	20.1 \pm 2.8	1.5 \pm 0.5	2.6 \pm 0.8	5.0 \pm 1.2	140 \pm 53	667 \pm 365	577 \pm 246	29 \pm 10
Gorramendi	2008	18.6 \pm 4.1	8.9 \pm 2.2	10.0 \pm 2.9	1.3 \pm 0.3	21.7 \pm 2.5	1.4 \pm 0.3	2.8 \pm 0.9	7.2 \pm 1.9	125 \pm 27	871 \pm 195	417 \pm 138	33 \pm 10
Legate	1411	12.9 \pm 8.9	9.9 \pm 1.5	9.6 \pm 1.7	2.2 \pm 0.4	22.3 \pm 1.4	1.6 \pm 0.6	2.6 \pm 0.7	6.8 \pm 1.4	123 \pm 29	1378 \pm 445	493 \pm 195	28 \pm 9
Aldude	1614	16.1 \pm 4.0	9.6 \pm 1.1	6.2 \pm 1.0	2.1 \pm 0.2	20.9 \pm 2.5	1.4 \pm 0.3	2.4 \pm 0.7	4.5 \pm 0.9	83 \pm 31	544 \pm 364	162 \pm 37	22 \pm 9
Belate	1846	13.3 \pm 4.4	12.2 \pm 2.2	6.6 \pm 2.2	1.9 \pm 0.4	22.0 \pm 1.9	1.3 \pm 0.2	2.1 \pm 0.5	6.5 \pm 0.7	119 \pm 18	304 \pm 165	507 \pm 180	32 \pm 8
Orokieta	1560	14.6 \pm 6.3	9.7 \pm 2.1	7.4 \pm 2.3	1.3 \pm 0.3	18.6 \pm 3.0	1.5 \pm 0.5	2.4 \pm 0.8	6.3 \pm 1.4	105 \pm 25	372 \pm 166	433 \pm 164	24 \pm 6
Orreaga	1506	25.0 \pm 6.8	9.6 \pm 1.4	7.6 \pm 2.0	1.2 \pm 0.3	22.6 \pm 2.5	1.2 \pm 0.3	2.2 \pm 0.5	5.7 \pm 0.9	110 \pm 30	1020 \pm 417	272 \pm 89	24 \pm 7
Larra	1158	35.4 \pm 9.6	12.6 \pm 3.2	11.4 \pm 2.1	1.4 \pm 0.3	24.4 \pm 3.3	1.6 \pm 0.6	2.3 \pm 0.2	7.1 \pm 2.0	83 \pm 38	193 \pm 79	125 \pm 91	24 \pm 6
Belagoa	1158	18.9 \pm 3.5	17.4 \pm 2.8	10.0 \pm 2.7	1.1 \pm 0.2	21.5 \pm 1.9	1.4 \pm 0.2	2.3 \pm 0.6	5.9 \pm 1.2	127 \pm 37	127 \pm 90	153 \pm 72	34 \pm 5
Lazar	1270	20.4 \pm 5.0	12.2 \pm 2.0	9.7 \pm 2.2	1.3 \pm 0.4	20.6 \pm 1.8	1.2 \pm 0.4	2.3 \pm 0.5	5.5 \pm 0.6	127 \pm 27	453 \pm 193	125 \pm 55	25 \pm 6
Izalzu	1270	18.8 \pm 3.1	12.1 \pm 2.2	9.8 \pm 3.5	2.1 \pm 0.4	21.2 \pm 3.0	1.3 \pm 0.4	2.4 \pm 0.5	6.4 \pm 1.5	100 \pm 22	740 \pm 311	108 \pm 81	28 \pm 11
Ori	1158	21.8 \pm 6.4	12.4 \pm 2.4	8.2 \pm 1.9	2.0 \pm 0.6	19.9 \pm 2.4	1.3 \pm 0.2	2.3 \pm 0.5	5.8 \pm 1.0	92 \pm 21	1033 \pm 261	135 \pm 51	25 \pm 10
Abodi	1270	23.5 \pm 5.9	11.7 \pm 1.5	6.9 \pm 1.6	1.9 \pm 0.6	20.6 \pm 3.5	1.4 \pm 0.3	2.2 \pm 0.5	6.2 \pm 1.1	100 \pm 31	207 \pm 167	193 \pm 48	35 \pm 10
Irati	1422	20.0 \pm 3.9	10.9 \pm 2.5	9.1 \pm 3.9	1.7 \pm 0.4	20.3 \pm 3.4	1.4 \pm 0.2	2.4 \pm 0.6	4.8 \pm 1.3	109 \pm 30	243 \pm 208	136 \pm 44	32 \pm 6
Mean	1571	19.6 \pm 7.7	11.3 \pm 3.1	8.2 \pm 2.7	1.7 \pm 0.5	21.3 \pm 2.9	1.4 \pm 0.4	2.3 \pm 0.6	6.1 \pm 1.4	113 \pm 41	556 \pm 420	338 \pm 300	30 \pm 11
Reference*			4.0–8.0	5.0–10.0	1.0–1.5	18.0–25.0	1.0–1.7	1.3–2.0	6.0–14.0	200–2000	< 1000	< 100	20–80

as were S concentrations (with the exception of Goizueta). Magnesium concentrations in foliage in more than half the stands were also higher than referenced values. Only N and P concentrations were within the cited range in all the stands. Most of the stands showed K foliage concentrations within the cited range too, except the two eastern-most sampled stands (Larra and Belagoa) where a surplus was observed.

For microelement concentrations, only Zn concentrations were within the referenced range in all stands, although many of the microelements were close to the minimum. However, Na concentrations were higher than the maximum and Fe was lower than the minimum in all stands. Some of the Cu concentrations were lower than the referenced minimum, and even in those stands where they fell within the range, concentrations were close to the minimum. Most stands had Mn concentrations within the range shown in the cited literature (only the concentrations in Legate, Orreaga, and Ori were higher than the maximum).

Comparing the main macronutrient ratios with the literature data reported for beech by Flückiger and Braun (1999), most of the stands (mainly in those belonging to the basophilic vegetation series) had higher N/P values than the referenced maximum. Stands presenting N/Mg and N/K values outside the range were few (Table 2).

Table 2. Main macronutrient ratios (mean \pm S.D.) in beech stands

Stand	Macronutrient ratios		
	N/Mg	N/P	N/K
Uitzi	13.3 \pm 4.1	18.3 \pm 2.8	4.0 \pm 0.9
Goizueta	22.1 \pm 8.9	16.3 \pm 5.0	2.8 \pm 1.0
Arano	15.0 \pm 9.0	15.1 \pm 3.6	3.2 \pm 0.9
Otsondo	9.9 \pm 1.7	14.2 \pm 3.5	3.2 \pm 1.0
Gorramendi	18.0 \pm 5.0	16.2 \pm 4.3	2.3 \pm 0.5
Legate	10.3 \pm 2.1	16.0 \pm 4.9	2.4 \pm 0.5
Aldude	10.0 \pm 1.9	14.9 \pm 3.1	3.4 \pm 0.5
Belate	12.1 \pm 2.8	17.3 \pm 3.7	3.6 \pm 1.0
Orokieta	14.3 \pm 4.5	13.3 \pm 4.5	2.8 \pm 1.0
Orreaga	19.4 \pm 5.4	20.4 \pm 5.0	3.1 \pm 0.8
Larra	18.6 \pm 6.9	16.2 \pm 3.5	2.2 \pm 0.4
Belagoa	20.4 \pm 4.0	16.1 \pm 3.2	2.2 \pm 0.5
Lazar	17.5 \pm 4.8	19.5 \pm 5.9	2.2 \pm 0.6
Izalzu	10.9 \pm 3.3	16.9 \pm 4.0	2.4 \pm 0.9
Ori	10.7 \pm 3.1	15.3 \pm 1.5	2.5 \pm 0.6
Abodi	12.7 \pm 6.8	15.0 \pm 2.3	3.1 \pm 0.9
Irati	13.0 \pm 5.4	14.8 \pm 2.8	2.4 \pm 0.8
Mean	14.5 \pm 6.3	16.3 \pm 4.2	2.8 \pm 0.9
Reference	8.0–15.0	8.0–15.0	2.0–3.0

Table 3. Differences in defoliation (%) and foliage element concentrations (mean \pm S.D.) between vegetation series. 'a' Not significant, 'b' significant at the 95% probability level, 'c' significant at the 99% probability level

Vegetation series	Defoliation	Macronutrients (mg/g)						Microelements ($\mu\text{g/g}$)				
		Ca	K	Mg	N	P	S	Cu	Fe	Mn	Na	Zn
Acidophilic	17.5 \pm 7.2	10.3 \pm 2.7	7.5 \pm 2.3	1.7 \pm 0.5	21.4 \pm 2.7	1.4 \pm 0.4	2.3 \pm 0.7	6.2 \pm 1.5	118 \pm 46	647 \pm 430	475 \pm 322	31 \pm 12
Basophilic	22.5 \pm 7.6	12.8 \pm 3.1	9.2 \pm 2.9	1.6 \pm 0.6	21.1 \pm 3.0	1.3 \pm 0.3	2.3 \pm 0.5	5.9 \pm 1.4	106 \pm 33	425 \pm 370	140 \pm 67	29 \pm 9
Significance	c	c	c	a	a	a	a	a	b	c	c	a

As far as differences between vegetation series are concerned, Ca and K concentrations were significantly higher in forest stands belonging to the basophilic vegetation series, whereas acidophilic forest stands had higher Fe, Mn, and Na concentrations. Also, defoliation was significantly greater in basophilic stands than in acidophilic stands, although both values were within the same defoliation class (slight defoliation) (Table 3).

Correlations between foliage element concentrations, when present, were weak (Table 4), so Principal Components Analysis was performed in order to find groups of element concentrations that are somehow joined. VARIMAX

Table 4. Correlation degree (Pearson's r test) between foliage element concentrations

	Ca	Cu	K	Fe	Mg	Mn	N	Na	P	S	Zn
Ca	1.00										
Cu	0.05 ^a	1.00									
K	0.15 ^b	0.13 ^b	1.00								
Fe	-0.06 ^a	0.08 ^a	-0.03 ^a	1.00							
Mg	0.10 ^a	0.00 ^a	-0.24 ^c	-0.13 ^a	1.00						
Mn	-0.21 ^c	0.06 ^a	0.05 ^a	0.04 ^a	0.23 ^c	1.00					
N	0.05 ^a	0.30 ^c	0.22 ^c	0.03 ^a	-0.12 ^a	0.07 ^a	1.00				
Na	-0.09 ^a	0.16 ^b	-0.22 ^c	0.16 ^b	0.09 ^a	0.02 ^a	-0.06 ^a	1.00			
P	0.11 ^a	0.14 ^b	0.09 ^a	-0.08 ^a	0.03 ^a	0.05 ^a	0.24 ^c	0.05 ^a	1.00		
S	-0.13 ^a	0.09 ^a	0.03 ^a	0.07 ^a	0.11 ^a	0.04 ^a	-0.29 ^c	0.07 ^a	-0.18 ^c	1.00	
Zn	0.21 ^c	0.30 ^c	0.01 ^a	0.16 ^b	0.03 ^a	-0.08 ^a	0.22 ^c	0.39 ^c	0.15 ^b	-0.10 ^a	1.00

^aNot significant.

^bSignificant at the 95% probability level.

^cSignificant at the 99% probability level.

Table 5. Groups of foliage element concentrations obtained by Principal Components Analysis

	Component				
	1	2	3	4	5
Na	0.818				
Zn	0.736				
Fe	0.474				
S		-0.753			
N		0.704			
P		0.598			
Mn			0.749		
Ca			-0.718		
K				0.817	
Cu				0.522	
Mg					0.841

Table 6. Correlation degree (Pearson's *r* test) between geographic coordinates of the stands and foliage element concentrations

	Cu	Na	K	Ca	Mg	N	Zn	Fe	Mn	P	S
Longitude	-0.13 ^a	-0.61 ^c	0.35 ^c	0.40 ^c	-0.11 ^a	0.02 ^a	-0.24 ^c	-0.14 ^b	-0.12 ^a	-0.09 ^a	0.05 ^a
Latitude	0.15 ^b	0.67 ^c	-0.12 ^a	-0.26 ^c	0.11 ^a	0.00 ^a	0.23 ^c	0.18 ^c	0.26 ^c	0.18 ^c	0.14 ^b

^aNot significant.

^bSignificant at the 95% probability level.

^cSignificant at the 99% probability level.

Table 7. Significant correlation degree (Pearson's *r* test) between the variation in defoliation from 1995 to 1997 and the variation in foliage element concentrations and macronutrient ratios in the same period

	ΔCu	ΔK	ΔMn	ΔN	ΔNa	ΔCa:Mg	ΔK:Mg	ΔN:Mg
ΔDef	-0.21 ^a	-0.21 ^a	0.24 ^a	-0.23 ^a	-0.20 ^a	-0.26 ^a	-0.25 ^a	-0.20 ^a

^aSignificant at the 95% probability level.

rotation gives five groups: Na–Zn–Fe; S–N–P; Mn–Ca; K–Cu; and Mg alone (Table 5).

Finally, correlation degrees between geographic coordinates of sampled stands and element concentrations were also calculated. The similar distribution patterns shown by Na, Zn, and Fe concentrations, which decrease from NW to SE, are the most interesting results obtained (Table 6).

Results corresponding to each sampling year were used separately, in order to calculate correlation degrees using the variations of every parameter for each tree during the interval between samplings.

Weak correlation degrees between the variation in defoliation from 1995 to 1997 (ΔDef) and the variation in some element concentrations and macronutrient ratios in the same period were also obtained: a positive correlation degree between ΔDef and ΔMn was found, and negative correlation degrees between ΔDef and ΔCu, ΔK, ΔN, ΔNa, ΔCa/Mg, ΔK/Mg, ΔN/Mg were found (Table 7). Using factor analysis, ΔDef is situated in the same group as ΔCu, ΔNa and ΔN (Table 8).

Interesting, although weak, correlations between the variation in the amount of precipitation from 1995 to 1997 and the variation in P foliage concentration, N/P ratio, and defoliation in the same period were obtained (Table 9).

Table 8. Groups of variations of different parameters obtained by Principal Components Analysis

	Component		
	1	2	3
$\Delta N/Mg$	0.886		
$\Delta K/Mg$	0.847		0.412
$\Delta Ca/Mg$	0.784		
ΔDef		-0.669	
ΔCu		0.591	
ΔNa		0.579	
$\Delta Rainfall$		0.534	
ΔN		0.481	
ΔK			0.806
ΔMn			0.613

Table 9. Significant correlation degree (Pearson's r test) between the variation in yearly rainfall from 1995 to 1997 and the variation in different parameters in the same period

	ΔP	$\Delta N:P$	$\Delta Defoliation$
$\Delta Rainfall$	-0.32 ^b	0.26 ^a	-0.30 ^b

^aSignificant at the 95% probability level.

^bSignificant at the 99% probability level.

4. Discussion

As mentioned, most of the values for Ca, Mg, and S are well above the referenced data. Nevertheless, macronutrient concentrations are most probably determined by ecological and geological conditions (soil, bedrock, etc.). The possibility of an atmospheric contribution to these concentrations must be discounted as, for example, the lowest S values have been found in the stands of Goizueta and Uitzu, which are close to a paper mill. In all Navarre, SO₂ concentration in the troposphere is exceeded only in the vicinity of this paper mill.

Calcium concentrations clearly exceed referenced values, even in the acidophilic stands, which are characterized by Si-rich oligotrophic soils.

As far as microelement concentrations are concerned, the same explanation given for the distribution of macronutrients is valid for Mn concentration.

The excess of Na found in all the stands is due to the proximity of the Atlantic Ocean, as the correlation degrees between Na concentration and the geographic coordinates (location of the stands) show. A decreasing gradient from NW to SE is evident. The low values found for Fe, Zn, and Cu concentrations show that the sampled territory is not subject to heavy anthropogenic pollution. However, taking into account the prevailing direction of the winds in the area (NW component), the Pearson's test and factor analysis results for Na, Zn, and Fe, together with their identical geographical behavior could indicate a long-range transport of Zn and Fe (in small quantities) from emitter points situated in the Bay of Biscay area, as has already been reported (Santamaría and Martín, 1997, 1998). Nevertheless, as the acidophilic stands are located northwest of the basophilic ones, the possibility that soil-related differences may be responsible for the geographical behavior shown by Fe concentrations cannot be ruled out. However, taking into account that differences in Zn foliage concentrations between vegetation series are not significant, the above explanation is unlikely to be the reason for such a gradient in this case.

With reference to macronutrient ratios, their values can show if there are any disturbances in the nutritional status of trees (Fürst, 1992; Stefan, 1995). Most of the stands exceed the referenced maximum for some of the main macronutrient ratios, so these stands may be suffering nutritional imbalances. According to Flückiger and Braun (1998, 1999), these imbalances increase the susceptibility of beech trees to pathogens and sucking insects, more on trees growing in poor acid soils than on trees belonging to basophilic forests.

Regarding differences between vegetation series, they are mainly due to the very different ecological conditions in which they grow. Lower Ca and K concentrations in acidophilic stands are explained by the leaching of base cations from the clay particles in acid soils, whereas higher Mn and Fe concentrations can be explained by the greater mobility of both elements in acid soils (and subsequent greater uptake by trees). Besides, Mn-rich soils are more frequent in the area occupied by the acidophilic forests (Alonso, 1998). The fact that acidophilic forests are nearer to the sea explains the higher Na concentrations found in these stands.

Some of the correlations obtained do not agree with the interactions between elements known for plants in general (Shuman, 1994): although P-Cu, Zn; Mn-Mg; Cu-P, Zn; Fe-Zn; and P-Zn are known to be antagonistic pairs, we have obtained positive correlation coefficients between these pairs of elements. Some of our results also contradict results obtained in other studies on beech foliage: Bidló and Kovács (1998) found a negative correlation between P-Cu, and positive correlations between Ca-Mn and S-N. Our results are just the opposite. The reason for such contradictory results could be that ours were

unwashed samples, so that the measured concentrations are both internal and exogenous.

Several authors have stated that yearly rainfall is a predominant causal factor responsible for changes in beech crown density (tree vitality) between successive years (Neiryneck and Roskams, 1999). The correlation between the variation in the amount of rainfall and the variation in defoliation agrees with such findings. The drier the year preceding crown condition assessment, the worse the tree vitality of beech forests is. On the other hand, results suggest that increasing amounts of rainfall lead to increasing N/P ratio. Taking into account that most stands in this study exceed referenced values for this ratio, wetter years could lead to further nutritional imbalances in beech forests, also increasing the susceptibility of trees to pest and pathogen attacks (Flückiger and Braun, 1998, 1999).

The negative correlations found between the evolution of Cu, K, N, and Na concentrations, Ca/Mg, K/Mg, and N/Mg ratios, and the evolution of defoliation indicate that changes in these element concentrations and ratios can cause visible changes in beech tree vitality. According to factor analysis results, of them, all the changes in Cu and N concentrations are best related to the variation in defoliation. It suggests that these two could be the key elements involved in the health of the studied beech stands. Moreover, as an increase in the foliage N concentration improves beech tree vitality, it also suggests that beech forests in Navarre are not N-saturated yet. Therefore, it may mean that the literature data reported for N/X ($X = \text{Mg, P, K}$) ratios are not very useful as a reference in this study.

The relationship between the variation in Na concentration and in defoliation is probably indirect and due to the close association between the changes in the amount of rainfall and the changes in foliage Na concentration (most of the rainfall events in the studied area come from the Atlantic Ocean).

5. Conclusions

At present, we do not know if the deficits and excesses found for several element concentrations and ratios are causing nutritional imbalances or are simply typical values for beech forests in the studied area. The active and exclusive ability of beech trees to fit to particular ecological conditions is already well known (Heinze, 1998). Consequently, the evolution of these parameters over the next few years should be studied in order to obtain more conclusive information.

Even though the studied area is not polluted, there are signs of a possible long-range transport of anthropogenic pollutants to this area that indicate the need for follow-up action in the future.

References

- Alonso, J.I., 1998. Estudio del contenido y distribución de los metales pesados en suelos de Navarra: cadmio, cobre, manganeso, níquel, plomo y cinc. Doctoral thesis, Universidad de Navarra.
- Anonymous, 1994–1997. Coyuntura agraria. Departamento de Agricultura, Ganadería y Alimentación, Government of Navarre.
- Anonymous, 1997. Results of large-scale foliar chemistry surveys (survey 1995 and data from previous years). In: Forest Foliar Condition in Europe. UN/ECE, Austrian Federal Forest Research Centre.
- Bidló, A., Kovács, G., 1998. Investigations on nutrient content in beech (*Fagus sylvatica* L.) seedlings of various provenances. *Agrokémia És Talajtan* 47, 317–328.
- Brumme, R., Leimcke, U., Matzner, E., 1992. Interception and uptake of NH_4 and NO_3 from wet deposition by above-ground parts of young beech (*Fagus sylvatica* L.) trees. *Plant and Soil* 142, 273–279.
- Bussotti, F., Ferretti, M., Cenni, E., Grossoni, P., 1995. Monitoring of mineral nutrients and trace elements in broadleaves: a survey in Tuscany. In: Nutrient Uptake and Cycling in Forest Ecosystems. European Commission, Ecosystem Research Report 21, pp. 99–106.
- DeVries, W., Leeters, E.E.J.M., Hendriks, C.M.A., van Dobben, H., van den Burg, J., Boumans, L.J.M., 1995. Large scale impacts of acid deposition on forests and forest soils in the Netherlands. In: Heij, G.J., Erisman, J.W. (Eds.), *Acid Rain Research: Do We Have Enough Answers?*, pp. 261–277.
- Fischer, B., Schweingruber, F.H., Keller, T., 1993. Impact of emissions from a garbage incinerator in Switzerland on the radial increment of beech trees. *Dendrochronologia* 11, 153–158.
- Flückiger, W., Braun, S., 1998. Nitrogen deposition in Swiss forests and its possible relevance for leaf nutrient status, parasite attacks and soil acidification. *Environ. Pollut.* 102, 69–76.
- Flückiger, W., Braun, S., 1999. Nitrogen and its effect on growth, nutrient status and parasite attacks in beech and Norway spruce. *Water Air Soil Pollut.* 116, 99–110.
- Fürst, A., 1992. Die Bedeutung des Schwefel/Stickstoffverhältnisses für die Beurteilung des Ernährungszustandes von Fichten. *FBVA Berichte* 71, 51–54.
- Heinze, M., 1998. Nutrition of forest trees on gypsum sites. *Forstwiss. Centralbl.* 117 (5), 267–276.
- Innes, J.L., 1993. *Forest health: its assessment and status*. Wallingford, CAB International.
- Maňkovská, B., 1997. Concentrations of nutritional and trace elements in spruce and beech foliage as an environmental indicator in Slovakia. *Lesnictví-Forestry* 43, 117–124.
- Maňkovská, B., 1998. The chemical composition of spruce and beech foliage as an environmental indicator in Slovakia. *Chemosphere* 36, 949–953.
- Meiwes, K.J., Merino, A., Beese, F.O., 1998. Chemical composition of throughfall, soil water, leaves and leaf litter in a beech forest receiving long term application of ammonium sulphate. *Plant Soil* 201, 217–230.
- Neiryneck, J., Roskams, P., 1999. Relationships between crown condition of beech (*Fagus sylvatica* L.) and throughfall chemistry. *Water Air Soil Pollut.* 116, 389–394.
- Rivas-Martínez, S., Bascónes, J.C., Fernández-González, F., Loidi, J., 1991. Vegetación del Pirineo Occidental y Navarra. *Itinera Geobot.* 5, 1–26.
- Santamaría, J.M., Martín, A., 1997. Moss bags as biomonitors of heavy metal deposition in Navarre, Spain. *Toxicol. Environ. Chem.* 60, 65–73.
- Santamaría, J.M., Martín, A., 1998. Influence of air pollution on the nutritional status of Navarra's forests, Spain. *Chemosphere* 36 (4–5), 943–948.
- Shuman, L.M., 1994. Mineral nutrition. In: Wilkinson, R.E. (Ed.), *Plant-Environment Interactions*. Marcel Dekker, New York, pp. 149–182.

- Stefan, K., 1995. Schwefel- und Nährstoffversorgung der Fichtennadeln im Gleinalmgebiet Mitt Der Forstl. Bundesversuchsanstalt Wien 163/5, 53–126.
- Szarek, G., Braniewski, S., Chrzanowska, E., Rieger, R., Rutkowska, L., 1993. Nutrients and pollutants in forest vegetation of the Ratanica watershed (Carpathian foothills, Southern Poland). *Ekologia Polska* 41, 375–392.
- UN/ECE, 1997. Manual on Methodologies and Criteria for Harmonised Sampling, Assessment, Monitoring and Analysis of the Effects of Air Pollution on Forests. 4th Edition, Hamburg.