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Air pollution, precipitation chemistry and forest health in the Retezat Mountains, Southern Carpathians, Romania

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Retezat Mountains are characterized by relatively clean air, acidic precipitation, and healthy, well growing forests.

Abstract

In the Retezat Mountains concentrations of O_3, NO_2 and SO_2 in summer season 2000–2002 were low and below toxicity levels for forest trees. While NH_3 concentrations were low in 2000, the 2001 and 2002 concentrations were elevated indicating possibility for increased N deposition to forest stands. More than 90% of the rain events were acidic with pH values <5.5, contributing to increased acidity of soils. Crown condition of Norway spruce (*Picea abies*) and European beech (*Fagus sylvatica*) was good, however, defoliation described as $>25\%$ of foliage injured increased from 9.1% in 2000 to 16.1% in 2002. Drought that occurred in the southern Carpathians between fall 2000 and summer 2002 and frequent acidic rainfalls could cause the observed decline of forest condition. Both Norway spruce and European beech with higher defoliation had lower annual radial increments compared to the trees with low defoliation. Ambient O_3 levels found in the Retezat did not affect crown condition of Norway spruce or European beech.

Keywords: Ozone; Norway spruce; European beech; Throughfall; Crown condition; Nutrients; Growth

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1. Introduction

High levels of nitrogen (N) and sulfur (S) deposition and increasing concentrations of ozone (O_3) may have undesirable effects on forest ecosystems in parts of the Carpathian Mountains of Central Europe (Bytnerowicz et al., 2004). Ozone alone has caused serious damage to vegetation in large areas of North America (Krupa and Manning, 1988; US EPA, 1996; Krupa et al., 2001) and Europe (deVries et al., 2003) and may also increase phytotoxic effects of other air pollutants, especially S and N oxides (Tingey and Reinert, 1975). It is expected that background ambient O_3 levels will increase in the future (Brasseur et al., 2001). Elevated O_3 concentrations have been measured in various locations of the Carpathian Mountains (Bytnerowicz et al., 2002). Long-term monitoring of ambient O_3 and other pollutants and investigations of biological and ecological changes are needed to understand the nature of the problem and to predict future risks to forest ecosystems in the Carpathian Mountains.

Air pollutants may have direct effects on health of forests, biodiversity and ecosystem processes. They may also have indirect effects on forests by promoting secondary stresses such as bark beetle infestations or toxicity of heavy metals in soils. Air pollution composition and distribution vary significantly in time and in space due to changes of climate and human activities, as well as environmental and physiographic changes with elevation. This temporal and spatial variability is particularly difficult to quantify in complex mountainous terrain. Therefore, monitoring of air pollutants for better understanding of its ecological effects has to be designed as a long-term activity, and location of monitoring sites must be chosen carefully. This study was intended as a long-term investigation of the effects of air pollution on forest ecosystems in the Retezat National Park of the southern Carpathian Mountains in Romania. The Park represents important biological values for the Carpathian Mountains and has been recently been added to the UNESCO Man & Biosphere Reserves network.

The specific objectives for this 2000–2002 study were: (a) to characterize spatial and temporal distribution of selected air pollutants; (b) to determine acidity and chemical composition of precipitation and throughfall in selected forest stands; (c) to evaluate soil physical and chemical conditions of the studied forest stands; (d) to characterize nutritional status of the dominant tree species; (e) to evaluate forest health according to the ICP-Forest protocols; (f) to assess growth of trees from different defoliation classes.

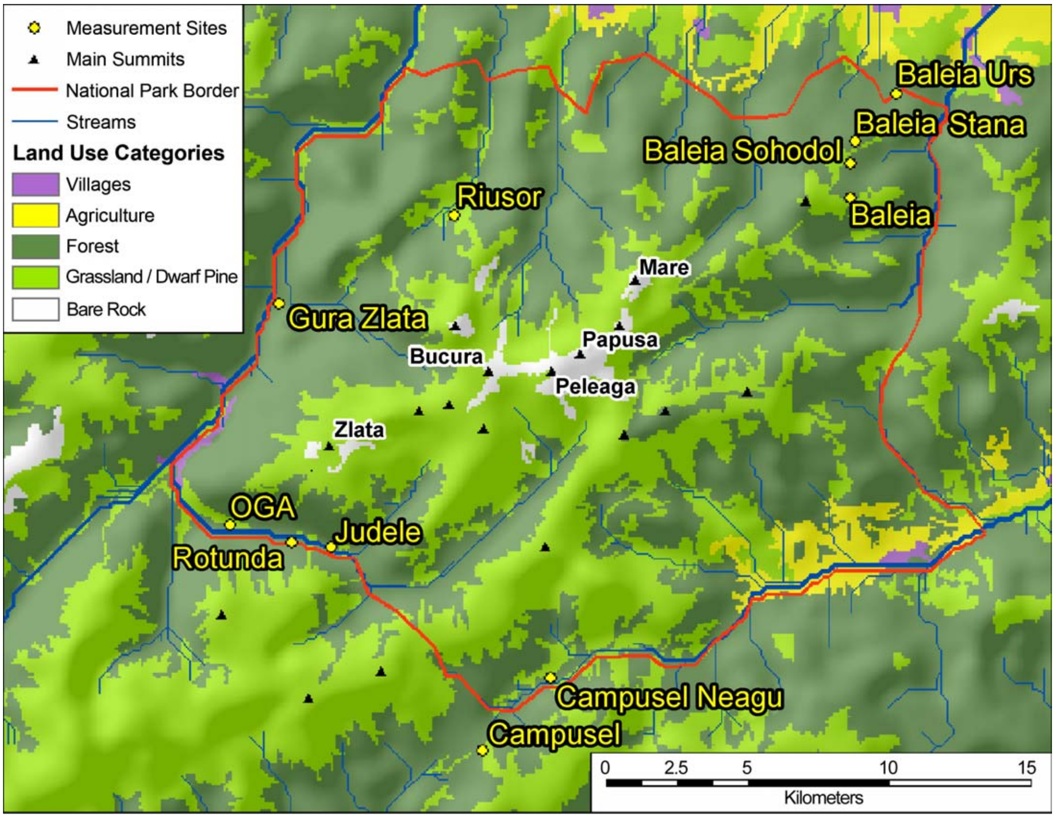


Fig. 1. Location of study sites in the Retezat Mountains (see Table 1 for detailed information on monitoring activities in each site).

Table 1
Research activities on the Retezat Mountains monitoring network

Site	Altitude (m)	Forest composition	Flora type	Air chemistry	Precipitation chemistry	Soil evaluation	Foliar analysis	Defoliation	Radial growth	Biodiversity
Judele	1190	*Picea abies*	*Oxalis*, *Dentaria*	+	+	+	+	+	+	+
Rotunda	1180	*Picea abies*, other conifers	*Asperula*, *Dentaria*	+		+	+	+	+	+
OGA	1000	*Fagus sylvatica*, *Picea abies*, *Abies alba*	*Asperula*, *Dentaria*	+		+		+	+	+
Gura Zlata	800	*Fagus sylvatica*, other broadleaves	*Luzula*, *Calamagrostis*	+	+	+	+	+	+	+
Riusor	1195	*Picea abies*, *Fagus sylvatica*, other broadleaves	*Poytrichum commune*	+		+	+	+	+	+
Baleia Sohodol	1300	*Picea abies*	*Oxalis*, *Dentaria*	+	+	+		+	+	+
Baleia Stana	1200	*Picea abies*, *Fraxinus*, other broadleaves	*Asperula*, *Dentaria*	+		+	+	+	+	+
Baleia Urs	800	*Fagus sylvatica*	*Asperula*, *Dentaria*	+		+	+	+	+	+
Baleia	1600			+						
Campusel Neagu	1100	*Picea abies*, *Fagus sylvatica*	*Luzula sylvatica*	+		+		+	+	+
Campusel	1400	*Fagus sylvatica*, *Picea abies*	*Asperula*, *Dentaria*	+	+	+	+	+	+	+

2. Study location

Retezat National Park is located in the southern Carpathian Mountains in Romania. The Park covers 80,000 ha and is in the highest range of the southern Carpathians with 19 peaks exceeding 2000-m elevation. These mountains are characterized by rich flora represented by 1186 vascular species, 104 sub-species and 312 varieties. The fauna is also rich and includes endangered species, such as chamois, bear, lynx, and

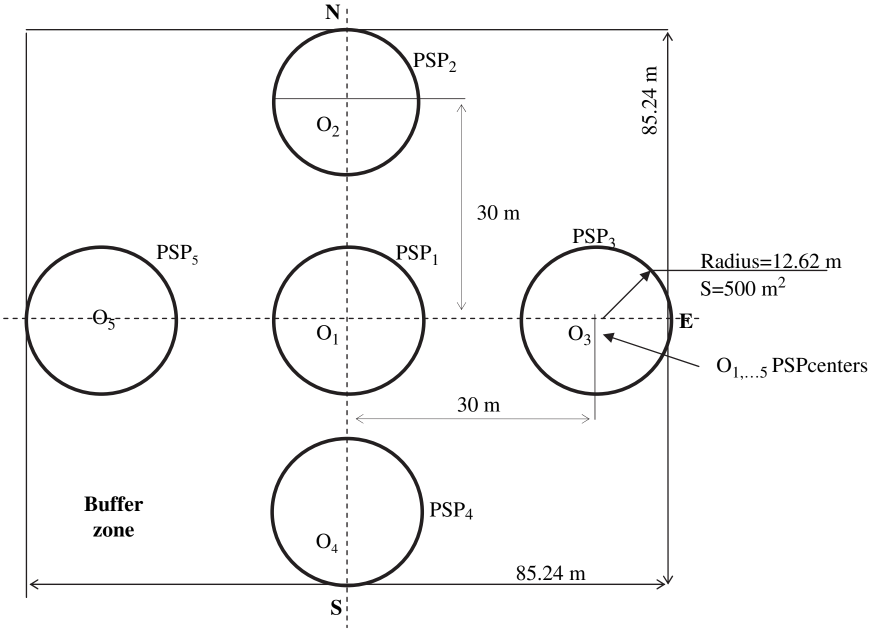


Fig. 2. Scheme of sample plot and spatial distribution of circular permanent sub-plots (PSPs) for tree evaluation used in the study.

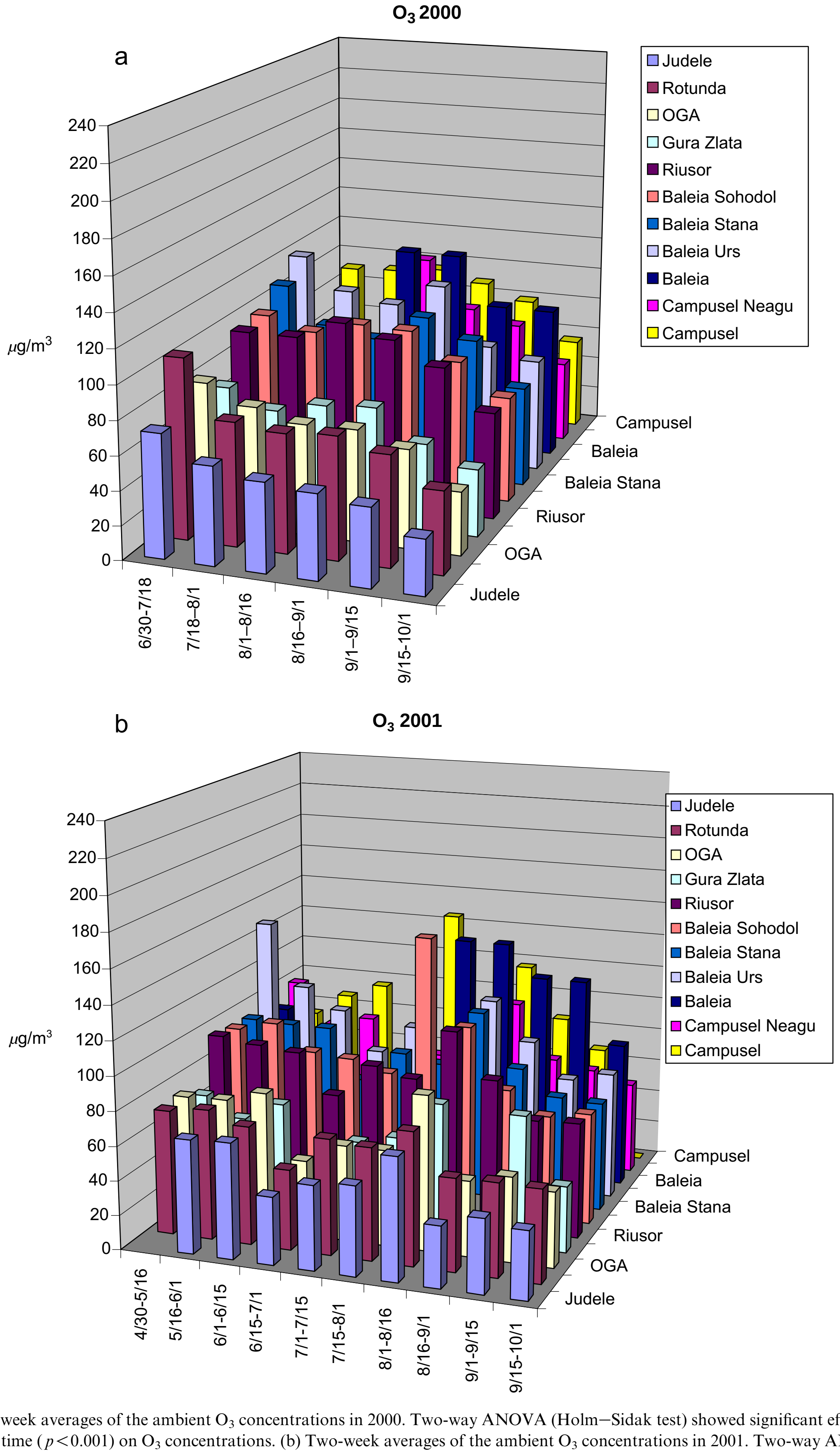


Fig. 3. (a) Two-week averages of the ambient O_3 concentrations in 2000. Two-way ANOVA (Holm–Sidak test) showed significant effects of location ($p<0.001$) and time ($p<0.001$) on O_3 concentrations. (b) Two-week averages of the ambient O_3 concentrations in 2001. Two-way ANOVA (Holm–Sidak test) showed significant effects of location ($p<0.001$) and time ($p<0.001$) on O_3 concentrations. (c) Two-week averages of the ambient O_3 concentrations in 2002. No analysis of variance was possible due to missing data.

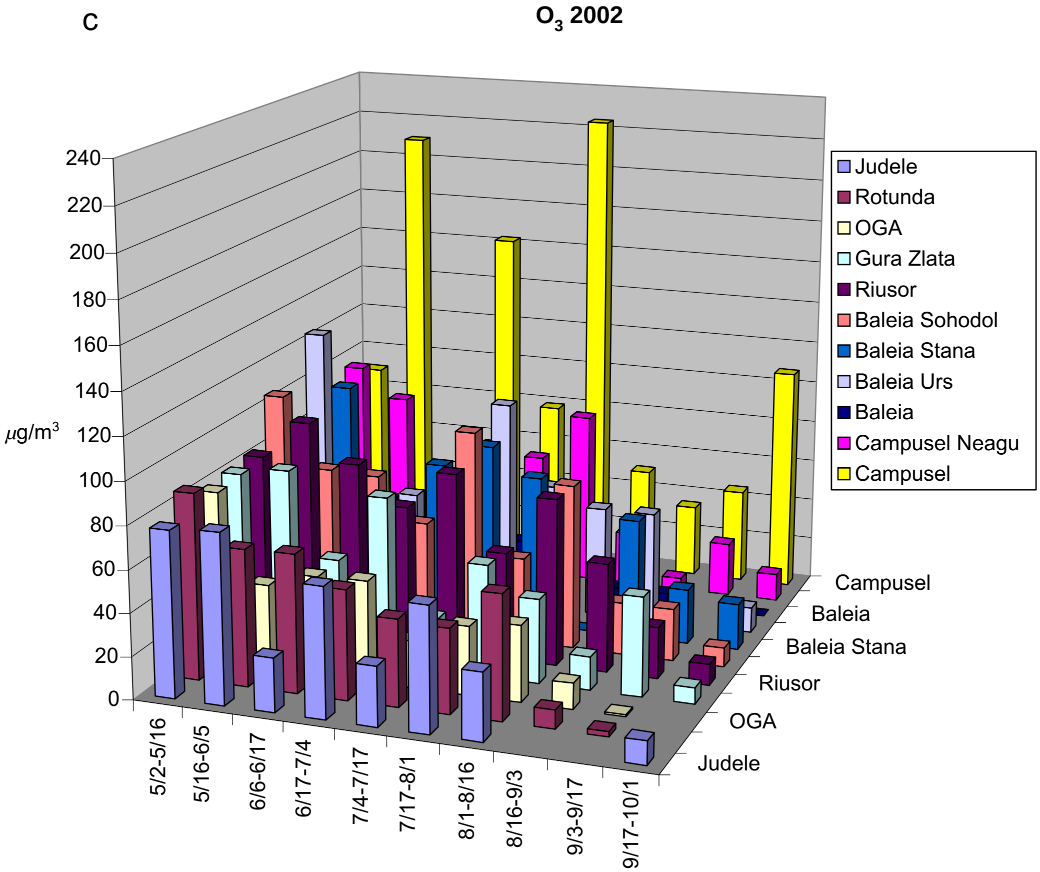


Fig. 3 (*continued*)

wolf. The Retezat National Park has been proposed to become a model for conservation efforts in Romania and other countries. The World Bank has supported this effort and the Retezat National Park has been included in the Global Environment Facility (GEF) network (Bytnerowicz et al., 2003).

A monitoring network consisting of 11 permanent sample plots was established for this study in the Retezat Mountains in early summer of 2000. These plots were located both within the Retezat National Park and outside of its boundaries in a forest zone at elevations ranging from 800 to 1500 m (Fig. 1 and Table 1). Monitoring sites were well exposed to the incoming air masses allowing for a good evaluation of the air pollution status and its effects on forest ecosystems.

3. Methodology

Concentrations of O_3, sulfur dioxide (SO_2), ammonia (NH_3), and nitrogen dioxide (NO_2) were monitored during the 2000–2002 growing seasons (May to October). Average 2-week long concentrations of air pollutants were monitored with Ogawa passive samplers for O_3 (Koutrakis et al., 1993), SO_2 and NO_2 (Ogawa & Co., USA, Inc., 1997), and with Gradko passive samplers for NH_3 (Hargreaves and Atkins, 1987). Passive samplers were collected from the field biweekly. Filters were sealed in plastic bottles and shipped to the USDA Forest Service Laboratory in Riverside, CA, for extraction and chemical analysis using ion chromatography (Dionex Model 4000i ion chromatograph) and colorimetry (TRAACS Technicon Autoanalyzer).

Bulk deposition (open-field and throughfall) was collected at four sites (Judele, Gura Zlata, Baleia Sohodol and Campusel). For each sample, pH, conductivity, concentration of nitrate (NO_3^-), sulfate (SO_4^{2-}), ammonium (NH_4^+), chloride (Cl^-) and selected cations (Na^+, Ca^{2+}, Mg^{2+} and K^+) were determined at the chemical laboratory of the Forest Research and Management Institute (Campulung Moldovenesc), in accordance with the methods recommended by the Internal Review of ICP Forests (2000).Precipitation samples were collected between 15 July and 16 September, on each year of the study (2000–2002). Samples were collected twice a month on the 16th and 30th/31st. All possible precautions were taken to

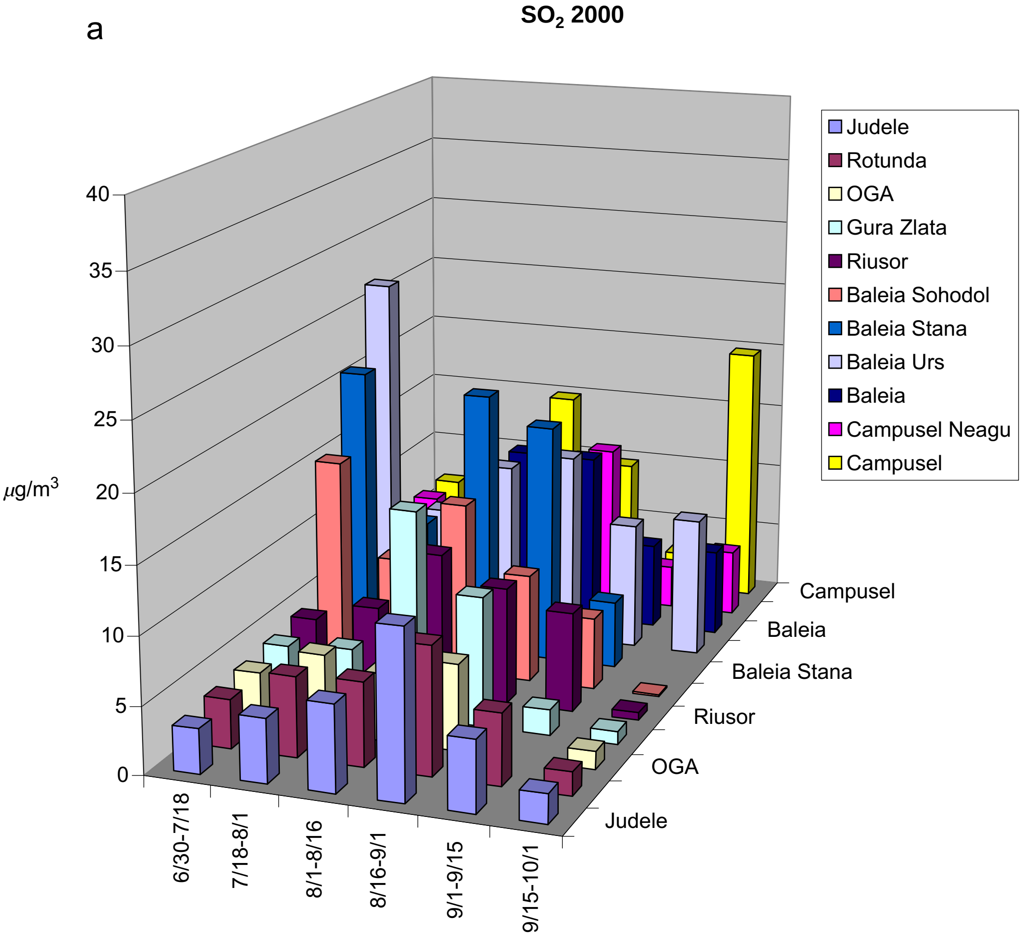


Fig. 4. (a) Two-week averages of the ambient SO_2 concentrations in 2000. Two-way ANOVA (Tukey test) showed significant effects of location ($p < 0.001$) and time ($p < 0.001$) on ozone concentrations. (b) Two-week averages of the ambient SO_2 concentrations in 2001. No analysis of variance was possible due to missing data. (c) Two-week averages of the ambient SO_2 concentrations in 2002. No analysis of variance was possible due to missing data.

minimize chemical contamination of the samples, especially from the surfaces of the samplers that were in contact with rain samples. After each collection, all equipment was rinsed with distilled water. Volume of collected samples was measured using a 250 ml graduated cylinder with a precision of ±2 ml (Barbu et al., 2000, 2001).

For the estimation of the input of the mineral ions in open-field and under the canopy the following formula was used:

$$Q_i = \sum_{j=1}^{n} \frac{P_j \cdot c_{ij}}{100}$$

where Q_i is the input of ion i (kg ha^{-1}) in j periods, P_j is the precipitation in the period j (L m^{-2}), and c_{ij} is the concentration of ion i in the period j (mg L^{-1}).

Soil samples were collected by genetic horizons in summer 2002 and analyzed at the Forest Research and Management Institute (Bucharest). Soil pH in water extracts was determined by the potentiometric method. Soil fine particles were divided into the following textural classes: sand (2–0.02 mm), silt (0.02–0.002 mm) and clay (<0.002 mm). The sand was fractioned by dry sieving, while the clay and silt fractions were determined by the pipette method (Barbu et al., 2000, 2001).

Foliage was collected for chemical foliar analysis for Norway spruce (*Picea abies*) needles from eight trees in three sites in March–April 2002 and for European beech (*Fagus sylvatica*) leaves from 18 trees in six research sites in August–September 2002. For each tree two branches were collected (with approx. 0.5 m length for spruce or about 30 leaves; approx. 50 g of dry substance, for beech). These branches were gathered for spruce from the upper third of the crown (the 7th verticillus and needle age of 1 and 2 years) and for beech from the upper part of the crown, from the middle and the upper part of the sprout. Concentration of sulfur (S) in needles and leaves was determined by digestion of samples in nitric acid (HNO_3) and perchloric acid ($HClO_4$) mixture and subsequent titration with barium

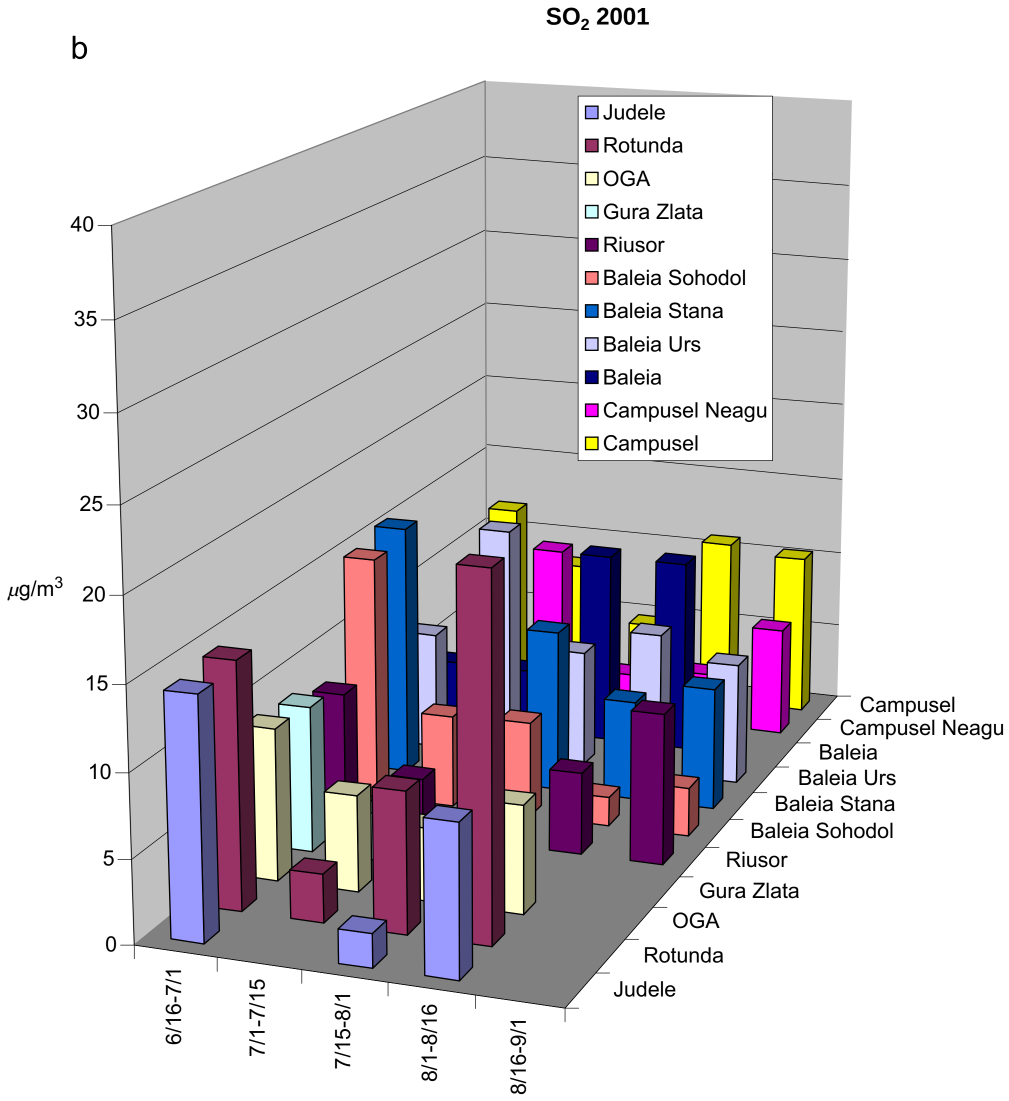


Fig. 4 (*continued*)

chloride ($BaCl_2$). Content of nitrogen (N) was determined by digestion (concentrated H_2SO_4 with K_2SO_4 and Se as catalysts), and the Kjeldahl method (distillation of NH_3 from the digested samples and titration of NH_4^+ with H_3BO_3). Phosphorus (P) was determined colorimetrically with molybdenum blue. Potassium (K) and calcium (Ca) were extracted by dry combustion (450 °C) followed by HCl (0.5 N) treatment and determined by flame photometry (Spectrometer UNICAM AAS 939).

An area of 0.7 ha was selected for intensive forest ecosystem studies at each research site. Each of these plots (Fig. 2) contained five clustered 500 m^2 area circular permanent sub-plots (PSP). Crown defoliation was assessed annually (July–August), in 5% increments at each study location (Internal Review of ICP Forests, 2000). Every year 1119 predominant, dominant and co-dominant (1, 2 and 3 Kraft classes) trees were assessed according to the EU-Scheme and ICP-Forests classification. Species composition of the understory vegetation was also described, using the Brown–Blanquet method. Biometric measurements of trees (species, diameter at breast height, tree height) were conducted. In addition, radial increment cores for the main species (spruce) were gathered with a Pressler borer from 16–20 selected trees located in the buffer zone of every PSP cluster (Fig. 2). In mixed stands (spruce and beech), 8–10 cores for each main species were gathered. For all sites studied 105 cores for spruce and 70 for beech were collected. Annual growth rings were measured and radial increment variation charts were developed for the defoliation class groups (0–1, crown defoliation ≤25%; and 2–3, crown defoliation over 25%), during the entire life of the selected trees.

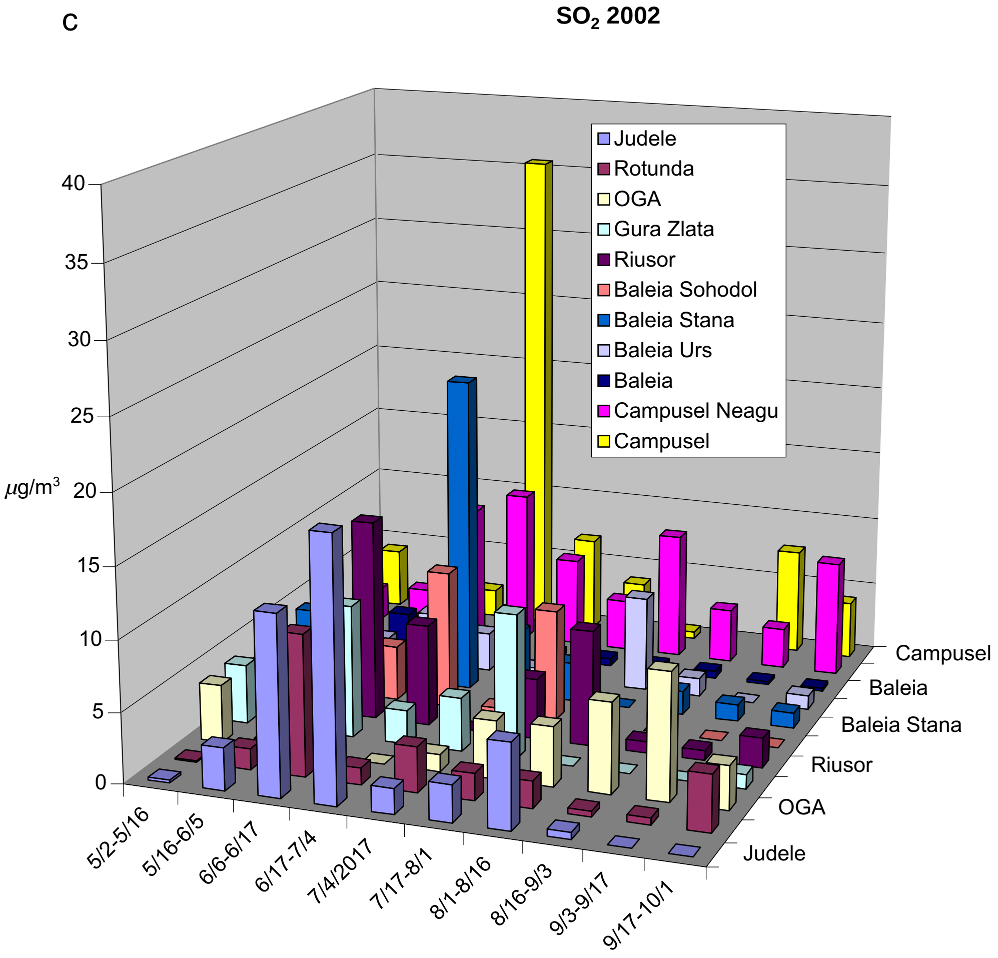


Fig. 4 (*continued*)

One- and two-way analysis of variance (ANOVA tests) were applied for statistical analysis of results (air chemistry, nutritional status, defoliation, annual radial growth). In several cases it was not possible to determine statistical significance for the above results due to missing data. Similarly, no statistical tests could be performed for precipitation chemistry and soil condition evaluation due to the small number of samples.

4. Results and discussion

4.1. Air chemistry

4.1.1. Two-week averages

Both spatial and temporal patterns of ambient O_3 distribution for the 2000–2001 seasons were similar (Fig. 3a,b). In 2002, until mid-August the patterns of O_3 distribution were similar to the 2000 and 2001 seasons, although some very high levels of O_3 were monitored on three occasions at the Campusel site. After mid-August ambient levels of O_3 dropped drastically (Fig. 3c). Ranges of the 2-week long average O_3 concentrations were similar for 2000 and 2001 (32–120 µg/m^3 and 35–142 µg/m^3, respectively), but not for 2002 (1–224 µg/m^3). With the exception of the Campusel site, during the 2002 season the O_3 concentrations determined were low and within ranges of values previously found in parts of the Romanian Carpathians and in the Sumava Mountains of the Czech Republic (Bytnerowicz et al., 2004). Such levels of O_3 are not considered toxic to most forest trees (Skärby and Karlsson, 1996), although effects on sensitive vegetation species cannot be ruled out.

Average SO_2 concentrations were quite variable in time and space without any clear patterns present (Fig. 4a–c). The concentration ranges were relatively low for 2000, 2001 and 2002 seasons (0.5–27 µg/m^3, 2–16 µg/m^3, and 0–36 µg/m^3, respectively). Such values are within ranges typical for this part of Europe (Kandler and Innes, 1995) and are not considered phytotoxic (Legge et al., 1998). The highest value for 2002 reflects one high reading for Campusel, with all other values for that year below 18 µg/m^3, similar to the values monitored the previous 2 years. Similarly, the high value for 2000 reflects one high reading for Baleia Urs, with all other values for that year below 20 μg/m^3.

Average NH_3 concentrations in 2000 were low and typical for remote locations (Fig. 5a). However, in 2001, and especially in 2002, highly elevated levels were occasionally monitored in some locations (Fig. 5b,c). The spikes in NH_3 concentrations were probably caused by some local emissions resulting from agricultural or forest operations (fires) or biological activity. Except for these rare spikes that reached over 110 μg/m^3, most (>90%) concentrations of NH_3 were below 20 μg/m^3. Although the monitored concentrations were well below phytotoxic levels (Bytnerowicz et al., 1998), they could contribute to elevated nitrogen deposition due to high deposition velocity of the pollutant (Hanson and Lindberg, 1991; Gessler and Rennenberg, 1998).

Concentrations of NO_2 in 2000 were low (below 5 μg/m^3) in most of the locations, however, occasionally elevated levels (as high as 28 μg/m^3) occurred in some locations (Gura Zlata, Baleia Sohodol, and Campusel) (Fig. 6a). In 2001, all locations were characterized by low levels (below 3 μg/m^3) of NO_2 (Fig. 6b). In 2002, increased levels of NO_2 were seen on several occasions in various locations (Fig. 6c). Such increased levels of the pollutant could be caused by local emissions from agricultural or forestry operations (burning of biomass, use of motor vehicles or other machinery, etc.). In general, even the highest levels of the pollutant observed (30 μg/m^3) were below toxicity levels of the pollutant (Bytnerowicz et al., 1998).

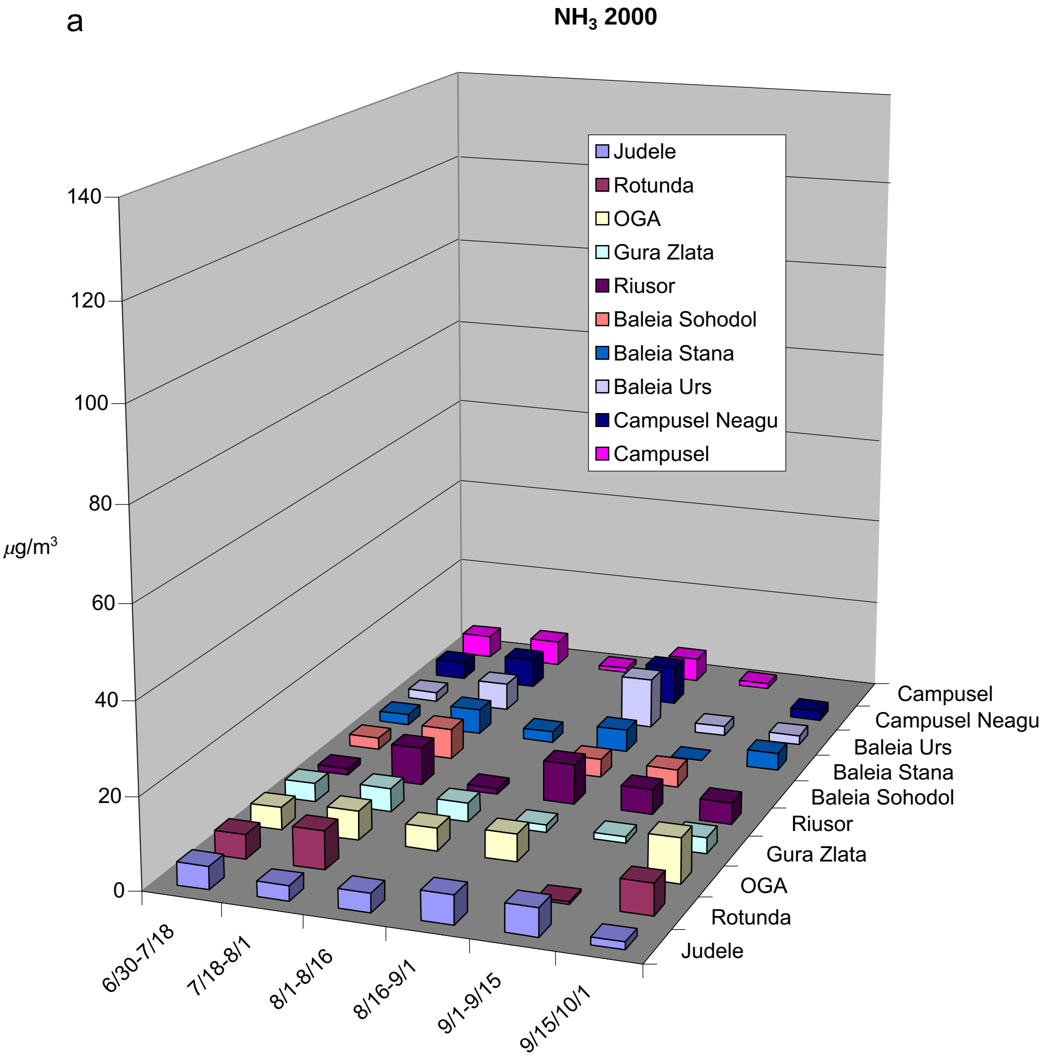
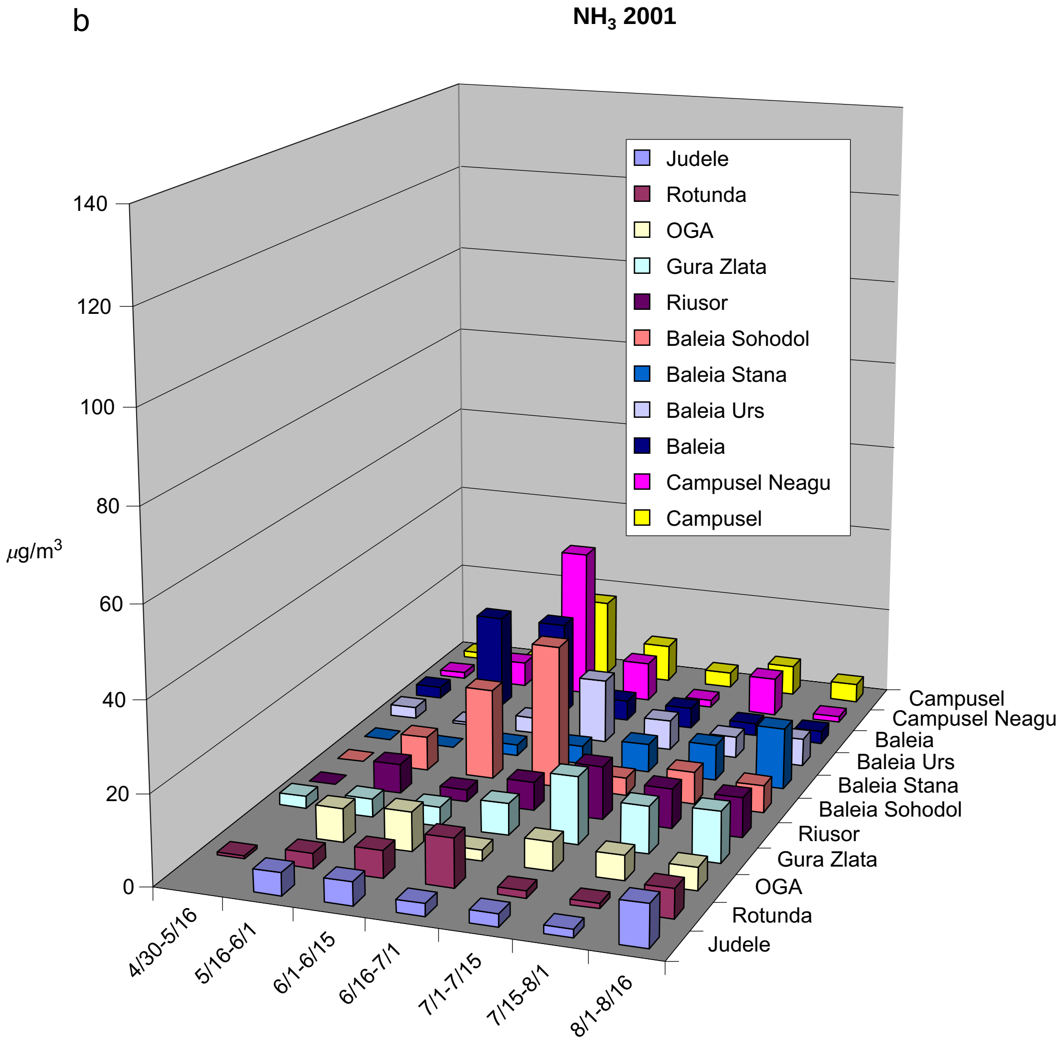


Fig. 5. (a) Two-week averages of the ambient NH_3 concentrations in 2000. No analysis of variance was possible due to missing data. (b) Two-week averages of the ambient NH_3 concentrations in 2001. No analysis of variance was possible due to missing data. (c) Two-week averages of the ambient NH_3 concentrations in 2002. Two-way ANOVA (Tukey test) showed no significant effects of locations ($p=0.651$) and time ($p=0.328$) on NH_3 concentrations.



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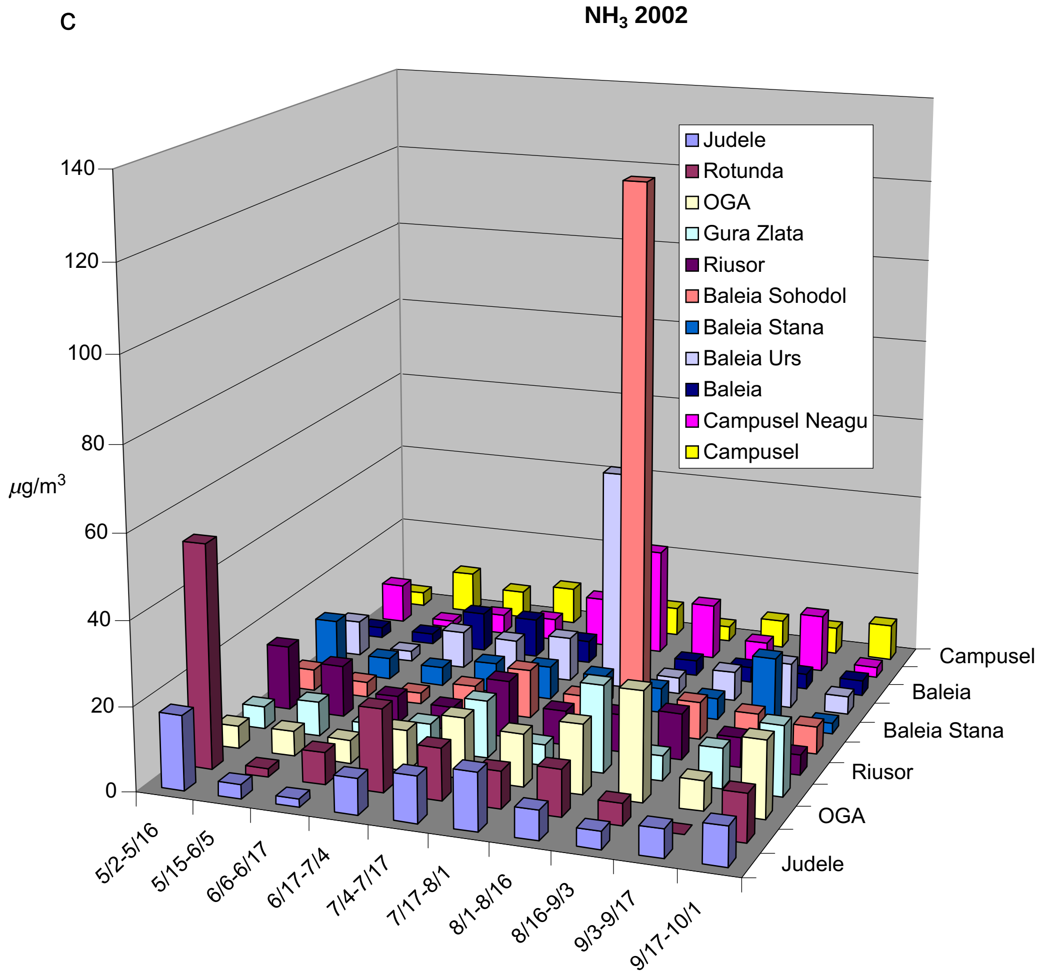
4.1.2. Seasonal means

Seasonal means for all four monitored pollutants for years 2000, 2001 and 2002 are presented in Fig. 7a–c, respectively. The 2000 and 2001 O_3 seasonal means were somewhat similar (ranges of 52–106 µg/m^3 and 51–96 µg/m^3, respectively) with lowest concentrations at Judele, Rotunda, OGA and Gura Zlata compared to the other sites. In 2002, seasonal mean concentrations were much lower (range for all sites except Campusel, 23–62 µg/m^3). It is interesting to note that the Baleia site experienced the highest levels in 2000 and 2001, but had the lowest seasonal mean in 2002. The recorded O_3 values are in agreement with ranges of seasonal means previously measured for the Romanian Carpathians in 1997–1999, and with the Brdy and Sumava Mountains of the Czech Republic (Bytnerowicz et al., 2004).

Seasonal SO_2 means for 2000 and 2001 seasons were similar (4–15 µg/m^3 and 5–15 µg/m^3, respectively), while the 2002 values were much lower (1–4 µg/m^3). No spatial patterns of SO_2 distribution were established. The recorded low SO_2 values are typical for the remote Central European locations (Kandler and Innes, 1995).

In general, seasonal means of NH_3 concentrations were lower in 2000 than in 2001 and especially 2002. Nevertheless, these monitored concentrations were much higher than levels found in pristine locations of the Canadian Rocky Mountains (Legge and Krupa, 1989) and eastern Sierra Nevada, California (Bytnerowicz and Fenn, 1996) or mountain locations affected by photochemical smog in southern California (Grosjean and Bytnerowicz, 1993).

Seasonal mean NO_2 concentrations during all 3 years of investigation were low (Bytnerowicz et al., 1998). The monitored levels were similar to those found at the Brenna site of the Polish Silesian Beskid Mountains (Bytnerowicz et al., 1999) and also at the Sequoia National Park, California, forest locations (Bytnerowicz et al., 2002). These levels are much lower than those



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occurring near urban agglomerations (Bytnerowicz et al., 1998).

4.2. Precipitation chemistry

Average precipitation quantities collected between July 15 and September 16 in 2000–2002 at the studied sites and the corresponding canopy interception are presented in Table 2. Mean interception in the canopy has different values depending on the density of the stand and the wind speed. The highest interception (29%) was recorded in the coniferous stand in Judele and the lowest (15%) in the beech mixed with other broadleaved species stand in Gura Zlata. Over the period 2000–2002, the average frequencies of rainfall with $pH < 5.0$ were higher in the open field (64–72%) than those registered under the canopy (39–67%) (Figs. 8 and 9). Altogether, in all sites the average frequencies of rain events with $pH < 5.5$ were about 90%. Under the canopy, frequencies of throughfall with $pH < 5.5$ were 37%. High frequencies of acid rain might have a negative effect on crown condition and health of trees. In addition, the high level of acid rain frequency affects the soil condition with subsequent negative influence on physiological processes of trees and forest stands (Edzards et al., 1997). In comparison with the frequency of acid rain events in previous years in Romanian monitoring plots for atmospheric deposition (Barbu et al., 2000, 2001) the frequency of acid rain events in the Retezat Mountains is about 2–2.5 higher. These results are preliminary and give only a preliminary indication of the variability of deposition in the Retezat Mountains.

Acidity of bulk precipitation was generally higher (lower pH values) than that of throughfall (Tables 3 and 4, respectively). At the same time, concentrations of ions in bulk precipitation were lower than those in throughfall. Analyzing the data from Tables 3 and 4, comparing concentrations of acidic (N, S) and alkaline (Ca, Mg) elements, as well as high values of water conductivity (salt accumulation, alkaline effect), it can be concluded that bulk precipitation in the Retezat Mountains was generally acidic. Therefore, rain precipitations could

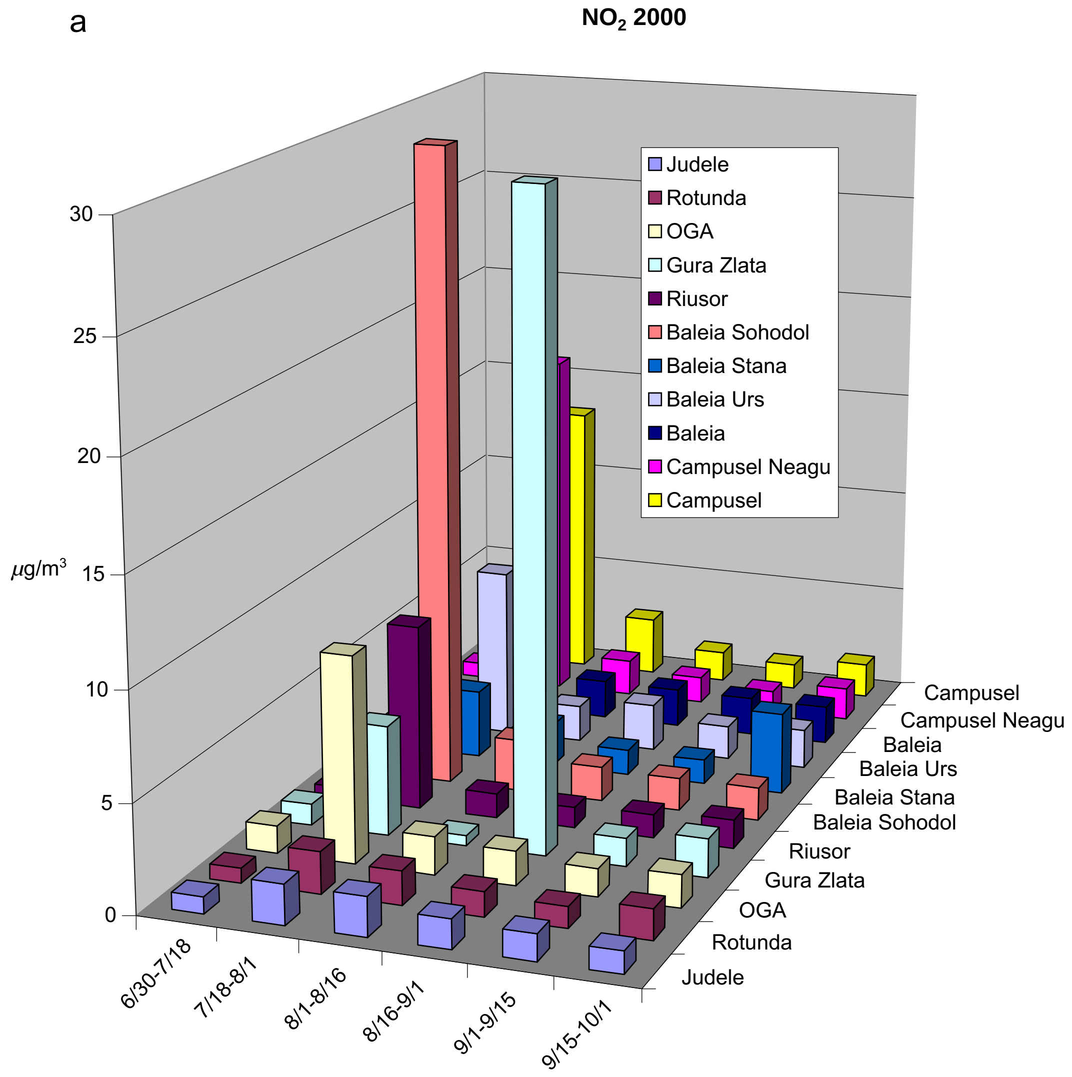


Fig. 6. (a) Two-week averages of the ambient NO_2 concentrations in 2000. Two-way ANOVA (Tukey test) showed no significant effects of locations ($p = 0.634$) and significant effect of time ($p = 0.004$) on NO_2 concentrations. (b) Two-week averages of the ambient NO_2 concentrations in 2001. Two-way ANOVA (Tukey test) showed no significant effects of locations ($p = 0.775$) and significant effects of time ($p < 0.001$) on NO_2 concentrations. (c) Two-week averages of the ambient NO_2 concentrations in 2002. No analysis of variance was possible due to missing data.

contribute to the continuous acidification processes of forest soil and negatively affect health of forest trees.

4.3. Soil condition

Soils in Retezat Mountains can be included in the following types and sub-types: Dystrudepts (Rotunda, Balea Stana, Balea Urs, Campusel Neagu), Dystrudepts, transition to Humic Dystrudepts (Gura Zlata, Riusor), Humicryols (Baleia Sohodol and Campusel), Humicryols transition to Haplorthods (OGA) and Haplorthods transition to Haplochyods (Judele). The soils have developed on parent materials mainly of acid character, rarely intermediate (gneiss, siliceous schist) on hard terrain specific to the mountain area.

Organic horizon (O) was composed of three sub-horizons the thickness of which depends on the intensity of the humification process. Therefore, the litter sub-horizon (LO) has a thickness between 0.5 and 3.0 cm, fermentation sub-horizon (FO) between 0.5 and 3.0 cm and humus sub-horizon (HO) between 0.0 and 1.5 cm.

The rock content (25–60%) on the soil profile was high, especially under the depth of 25–30 cm, and at depth of 50–140 cm the compact parental material of soils starts. The rock content and the compact parental material influenced both the edafic volume of soil (sub-medium (0.43 m^3/m^2) to high (0.76 m^3/m^2)) and its physiological and morphological thickness.

Texture of soils was generally loamy-sandy, rarely sandy-loamy, loamy-loam-sandy and the soil structure

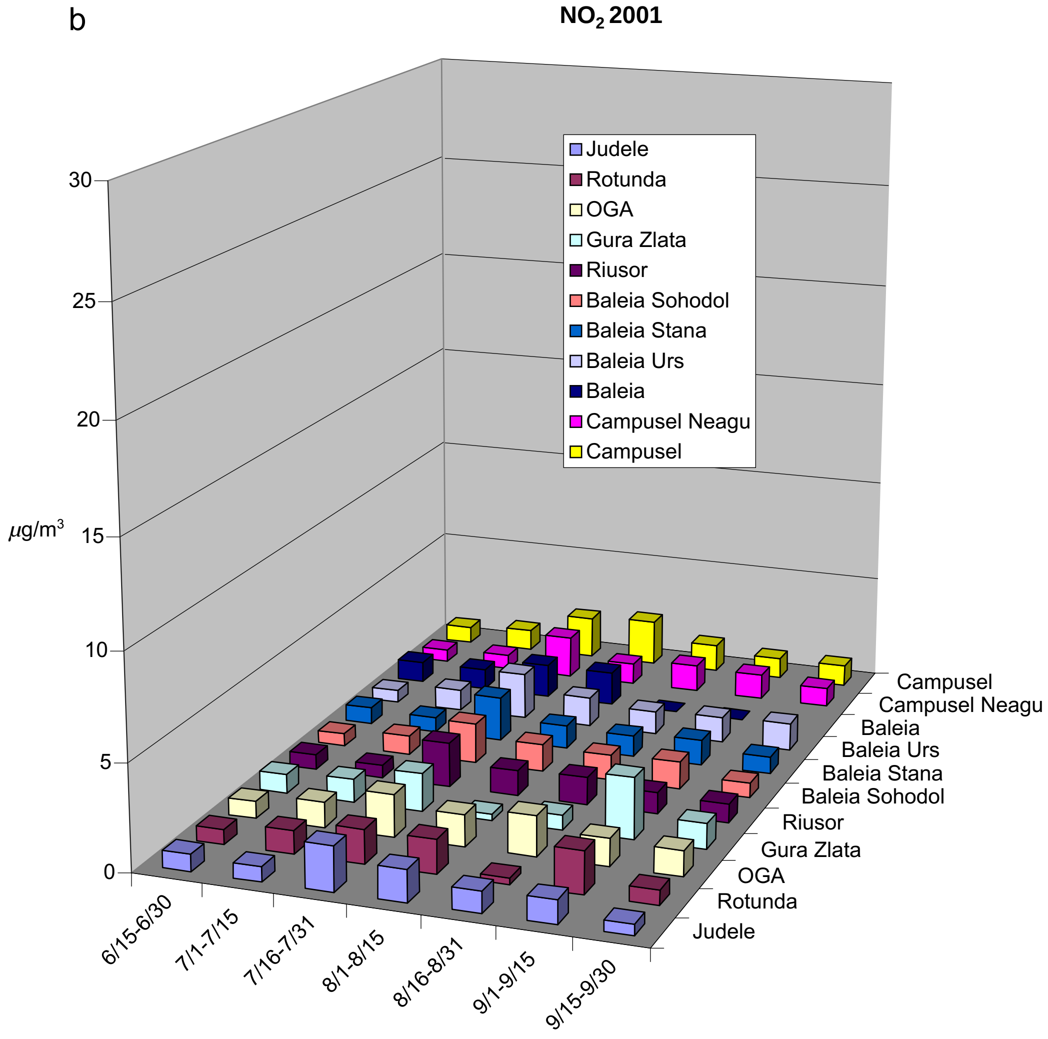


Fig. 6 (*continued*)

was polyhedron sub-angular small-very small (rarely medium), little or at most moderately developed. The Retezat soils were very acidic in upper (15–25 cm) horizons (pH 3.4–4.2) or even on the whole profile with pH 3.4–4.2 (OGA and Baleia Sohodol). An exception is the Baleia Urs location where pH values were between 5.1 and 5.2 (moderate acid soil) on almost the entire profile. Under the depth of 15–25 cm to parental material the soils were severely acidic (pH 4.3–5.0), except Baleia Urs (pH 5.1–5.2), Rotunda (pH 5.2–5.4) and Gura Zlata (pH 5.2–5.4). The very strong acidic characteristic in the upper horizons of the soils can be partially attributed to atmospheric deposition of S and N as SO_4^{2-}, NO_3^- and NH_4^+ in wet precipitation and dry deposition of SO_2 and NO_2. Although acidity of soils is common for these types of geological material, it is possible that the long-term SO_2 and NO_x deposition could further increase soil acidity in the Retezat Mountains.

Taking into account the granitic bedrock of the Retezat Mountains with low capacity for buffering the protons, the region is sensitive to any acidic input and soil acidification could occur in the future. Therefore, long-term monitoring of deposition and soil chemistry is necessary to provide documentation that this insidious process may actually be occurring.

The estimated levels of N and S inputs for the growing period were 2–4 kg/ha and for the entire year about 4–6 kg/ha. According to the European scale for the evaluation of the intensity of S-SO_4 and N-NO_3+N-NH_4 deposition, these values express “very low-low” levels of deposition (PCC West, 1994). These current levels of S and N deposition should not create acidification problems for the soil and trees (Barbu, 1991).

4.4. Nutritional status of trees

Concentrations of N, Ca, K, P and S in beech foliage (Fig. 10) were generally within the ranges considered as

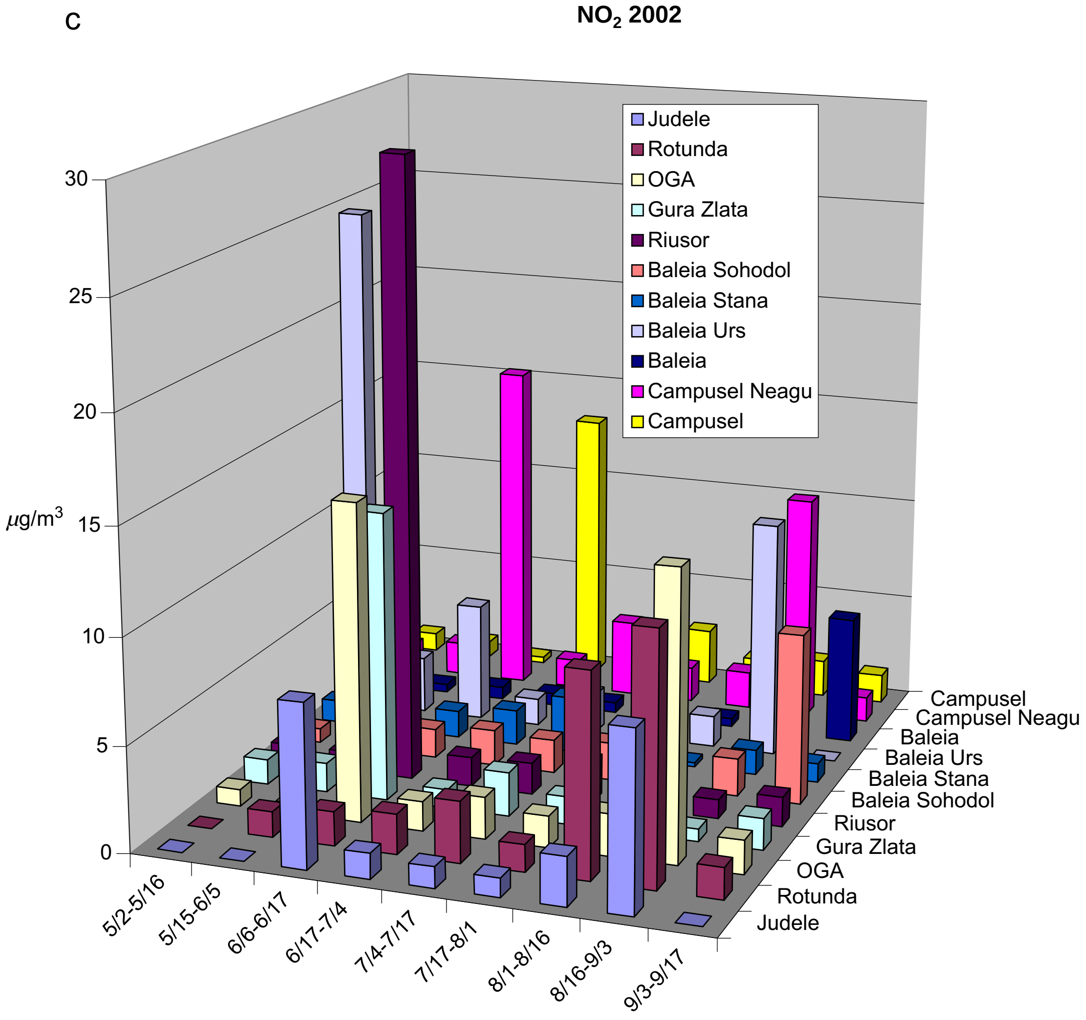


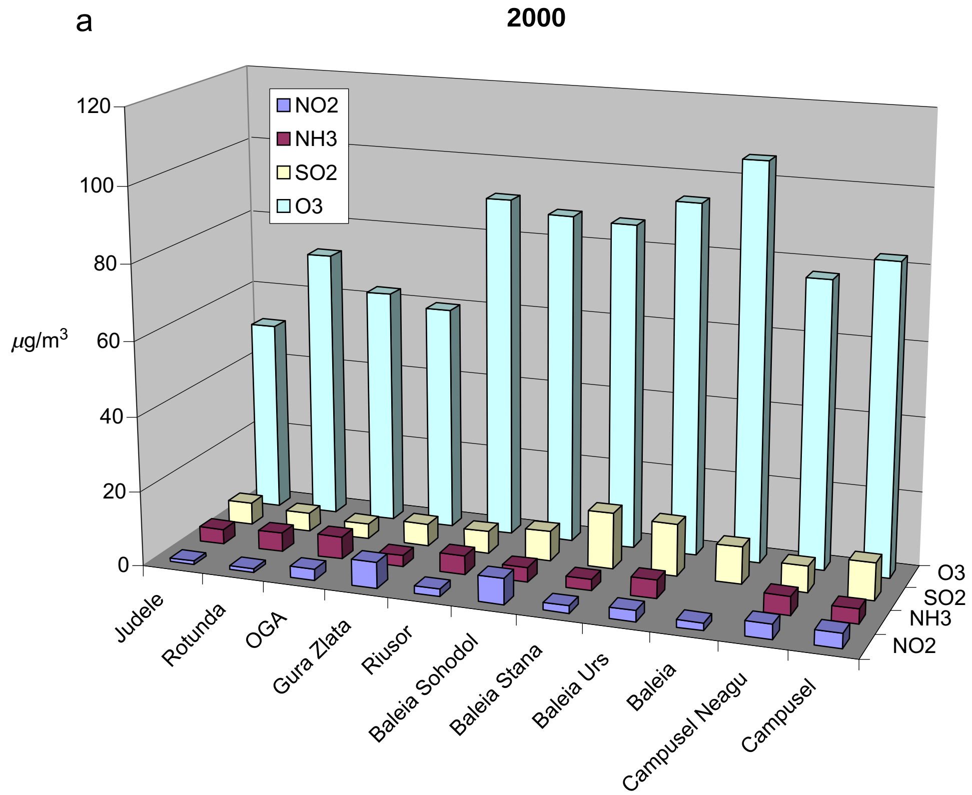
Fig. 6 (*continued*)

normal for broadleaf trees (Kramer and Kozlowski, 1979; Mankovska, 1997). In Baleia Stana, S concentrations were higher than in other sites, and at the levels considered high for this species (Stefan et al., 1997). In spruce foliage (Fig. 11), concentrations of N, Ca, K and P were within normal physiological levels for that species while the S concentrations were low (Stefan et al., 1997).

4.5. Defoliation

Some significant differences in the occurrence of high levels of defoliation (Classes 2–4, defoliation >25%) were noticed between the individual study sites and as well between the years (Fig. 12). Percentage of trees with defoliation >25% of all trees on all plots in 2000, 2001 and 2002 were 9.1%, 13.9%, and 16.1%, respectively. A possible increase of defoliation percentage of damaged trees (defoliation >25%) with time was seen both for spruce and beech, although the statistical significance of this apparent increase was not confirmed due to the very short period of time (2000–2002) when the assessment was made (Fig. 13). The worsening of health status in 2001 and 2002 compared with 2000 for beech can be explained by an excessive drought that occurred throughout the entire Romanian Carpathian Mountains from autumn 2000 to summer 2002 and the high frequencies of highly acidic rainfalls. However, in general, condition of spruce and beech in the Retezat Mountains compared to their status in other European forests (Lorenz et al., 2003) was good. For the whole of Europe, the average percentage of trees with defoliation >25% ranged between 18% and 25.5% for beech and 32.8% and 34.1% for spruce for the same years (Lorenz et al., 2003).

There was no correlation between ambient O_3 (described as mean seasonal concentrations for every site) and percent of trees with >25% defoliation (Fig. 14). This was to be expected since the O_3 concentrations at Retezat Mountains were low and below the a



b

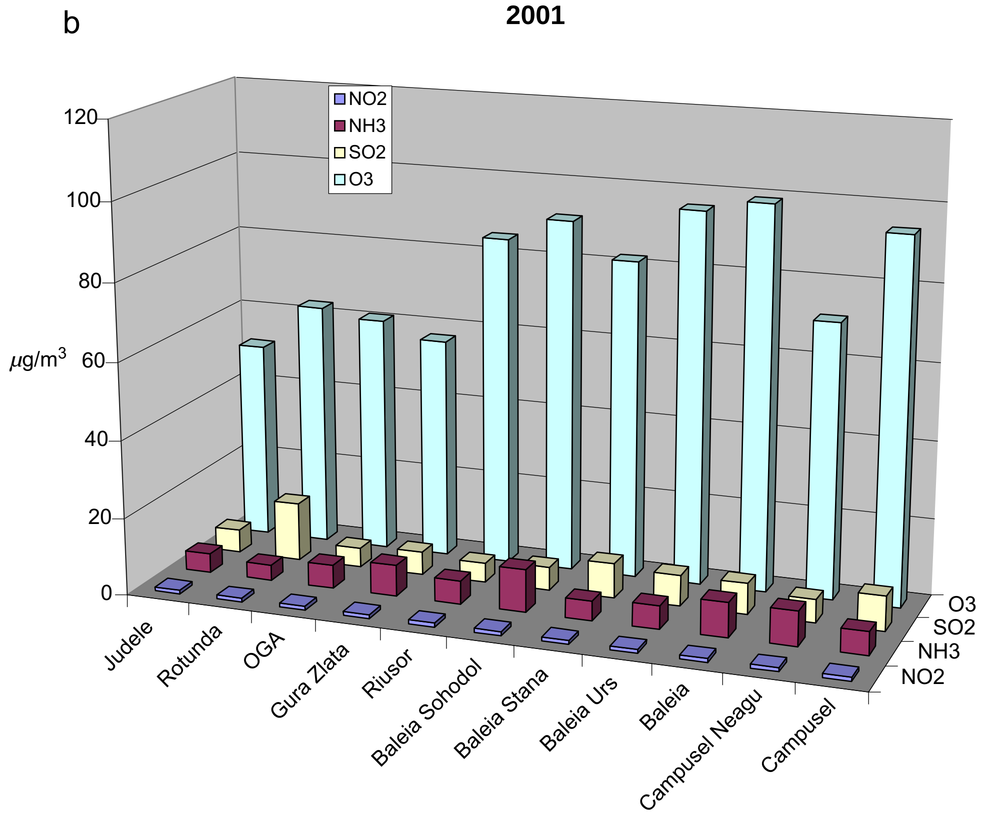


Fig. 7. (a) Seasonal average concentrations of air pollutants in 2000. (b) Seasonal average concentrations of air pollutants in 2001. (c) Seasonal average concentrations of air pollutants in 2002.

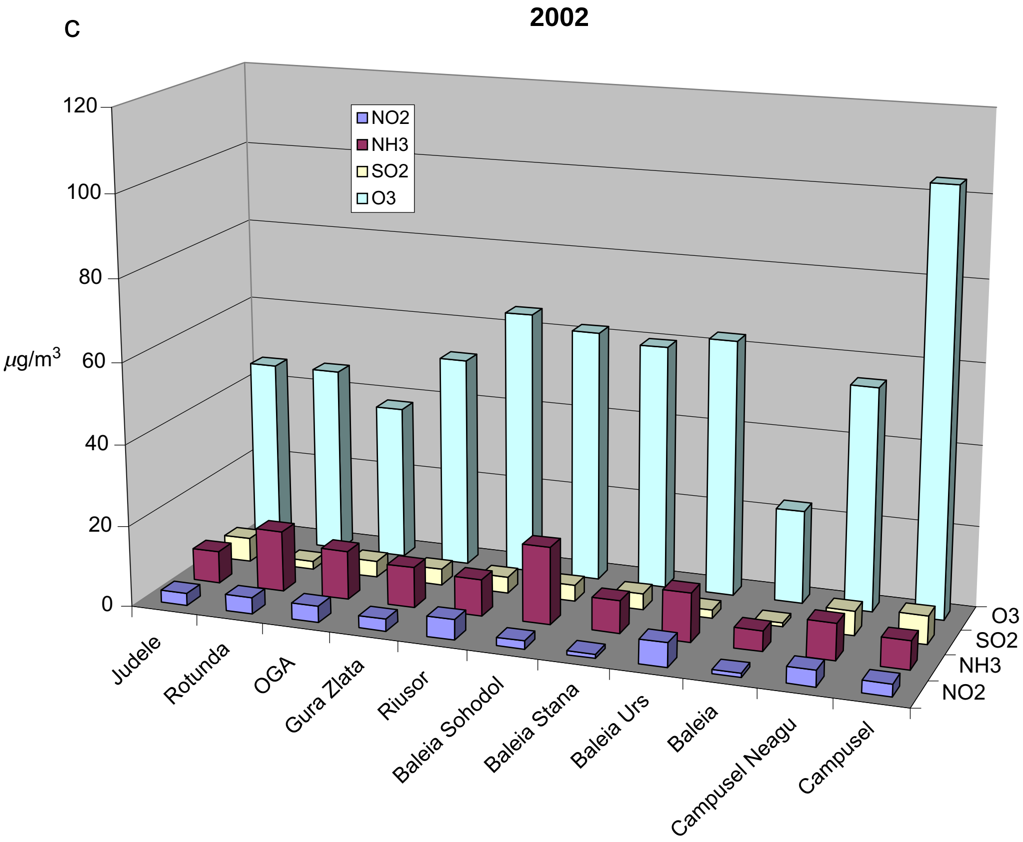


Fig. 7 (*continued*)

levels considered to be toxic for spruce (Wieser and Havranek, 1996) and beech (Skärby and Karlsson, 1996).

4.6. Biodiversity

A list for each species by layers (A, trees; B, shrubs; C, higher plants; D, mosses) has been established based on the biodiversity studies carried out in each site of the research network. The percentage coverage has been measured for each layer by using the Brown–Blanquet method. These data enabled identification of plant communities for each research site (Table 5).

The studied stands varied starting from pure spruce or beech forests to mixed forests with spruce, fir, beech and other conifers and broadleaved species. The number of plant species was different from one stand type to another varying from 10 to 35. The beech and mixed forests were richer in species, both in number and in value (*Hieracium*, *Calamagrostis*). Similarly, diameter and height of trees in the studied stands varied according to the stand type and structure (Table 6).

4.7. Radial increments

The average annual increment (the average width of the annual ring) was measured for defoliation classes 0–1 and 2–3 groups during the entire life of the selected trees. Results from the Riusor site for beech

Table 2
Average quantities of rainwater (mm) measured in open field, under the canopy and interception in the canopy for sites studied within 15 July–15 September over the period 2000–2002 (Retezat National Park, Romania)

Site	Average quantities of precipitation (mm)		Mean interception (%)
	Open field	Under canopy	
Judele	328	232	29
Gura Zlata	256	218	15
Baleia Sohodol	314	262	17
Campuşel	220	164	25

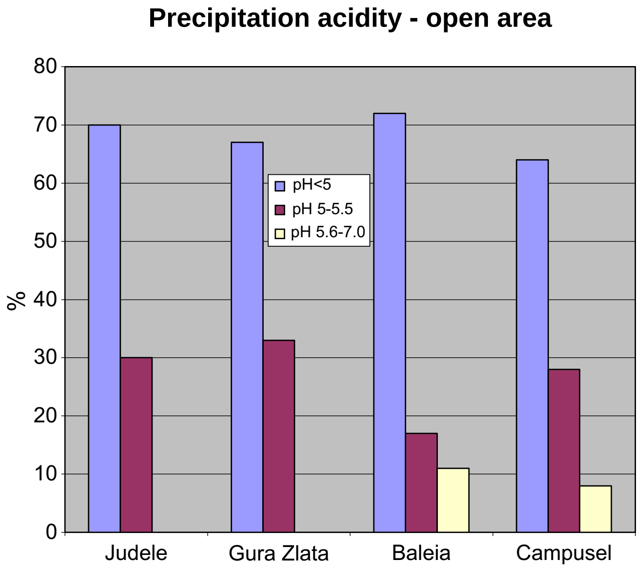


Fig. 8. Occurrence of rain events of various acidities during the 2000–2002 summer seasons. No statistical tests could be performed due to the small number of samples.

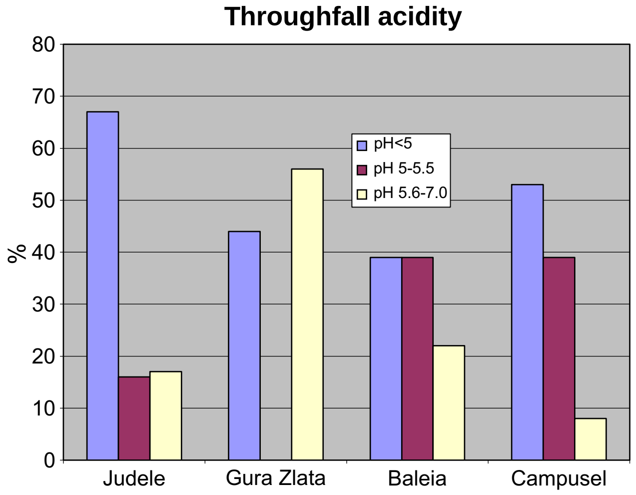


Fig. 9. Occurrence of throughfall of various acidities during the 2000–2002 summer seasons. No statistical tests could be performed due to the small number of samples.

(Fig. 15a) and spruce (Fig. 15b) were quite typical for most of the study sites showing significantly lower values for the damaged trees (classes 2–3) than for undamaged ones (classes 0–1). Average annual increments for beech (Fig. 16) and spruce (Fig. 17) indicated a trend towards lower growth of damaged trees (classes 2–3) versus undamaged ones (classes 0–1) in all measured plots. This may be explained by the fact that environmental factors that promote better growth of trees (such as availability of water and nutrients, good air quality, lack of pathogens and pests, etc.) also promote healthy trees with dense, healthy crowns (Edzards et al., 1997). Taking into account the species, age, altitude and productivity, the evaluated trees should be considered as healthy and well growing when compared with similar stands from other regions in Romania (Badea and Tanase, 2003).

Table 3
Average concentrations of ions in bulk precipitation samples collected during the period 2000–2002

Site	pH	Conductance (µS/cm)	S-SO_4 (mg/L)	Cl (mg/L)	N-NO_3 (mg/L)	N-NH_4 (mg/L)	Na (mg/L)	K (mg/L)	Mg (mg/L)	Ca (mg/L)
Judele	4.895	29.835	1.428	0.595	0.091	0.469	0.282	1.028	0.075	0.539
Gura Zlata	4.624	41.508	1.813	0.550	0.000	0.532	0.528	0.558	0.078	0.617
Baleia	4.956	28.871	1.876	0.649	0.052	0.417	0.420	0.400	0.248	1.234
Campusel	4.845	33.324	1.760	0.828	0.339	0.826	0.466	0.536	0.122	1.103

Table 4
Average concentrations of ions in throughfall samples collected during the period 2000–2002

Site	pH	Conductance (µS/cm)	S-SO_4 (mg/L)	Cl (mg/L)	N-NO_3 (mg/L)	N-NH_4 (mg/L)	Na (mg/L)	K (mg/L)	Mg (mg/L)	Ca (mg/L)
Judele	4.931	38.942	2.256	1.103	0.083	1.284	0.373	3.602	0.281	1.227
Gura Zlata	5.356	32.337	1.847	0.688	0.111	0.536	0.296	2.181	0.220	1.038
Baleia	5.194	39.289	2.179	0.886	0.198	1.170	0.298	2.330	0.228	1.474
Campusel	5.131	38.243	2.312	0.828	0.163	0.971	0.314	2.454	0.209	1.227

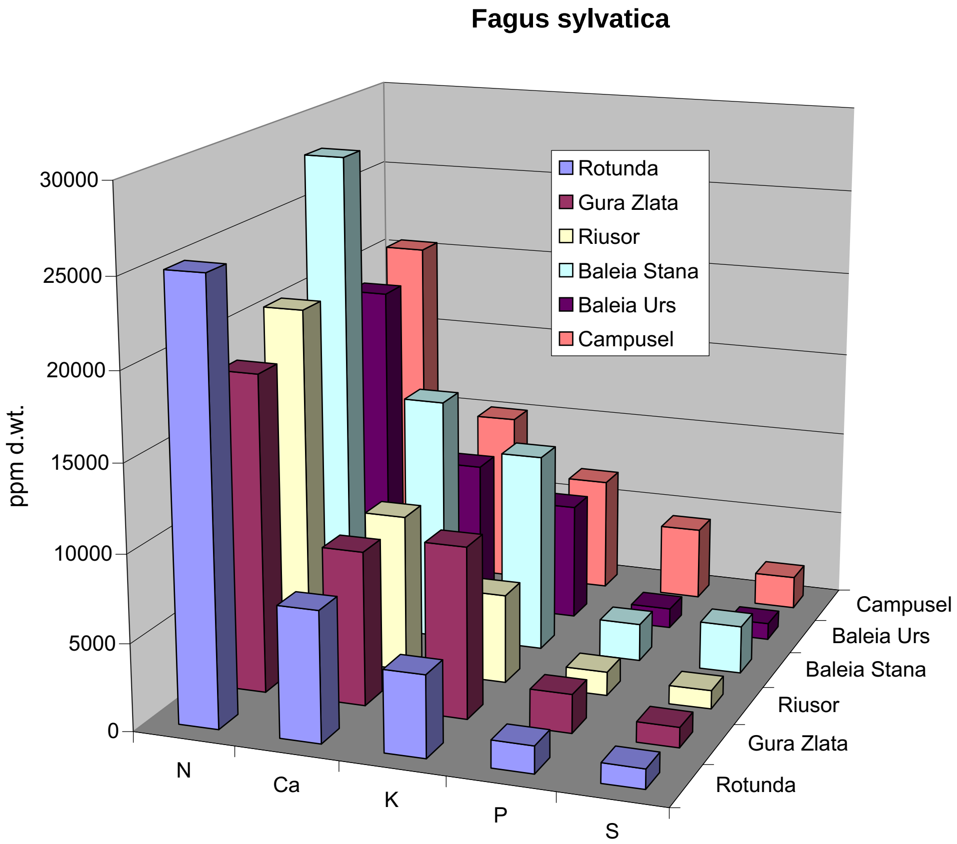


Fig. 10. Concentration of major nutrients in foliage of *Fagus sylvatica*. Two-way ANOVA indicated no significant differences between the study sites for individual nutrients ($F = 1.04 < F_{5\%} = 4.50$).

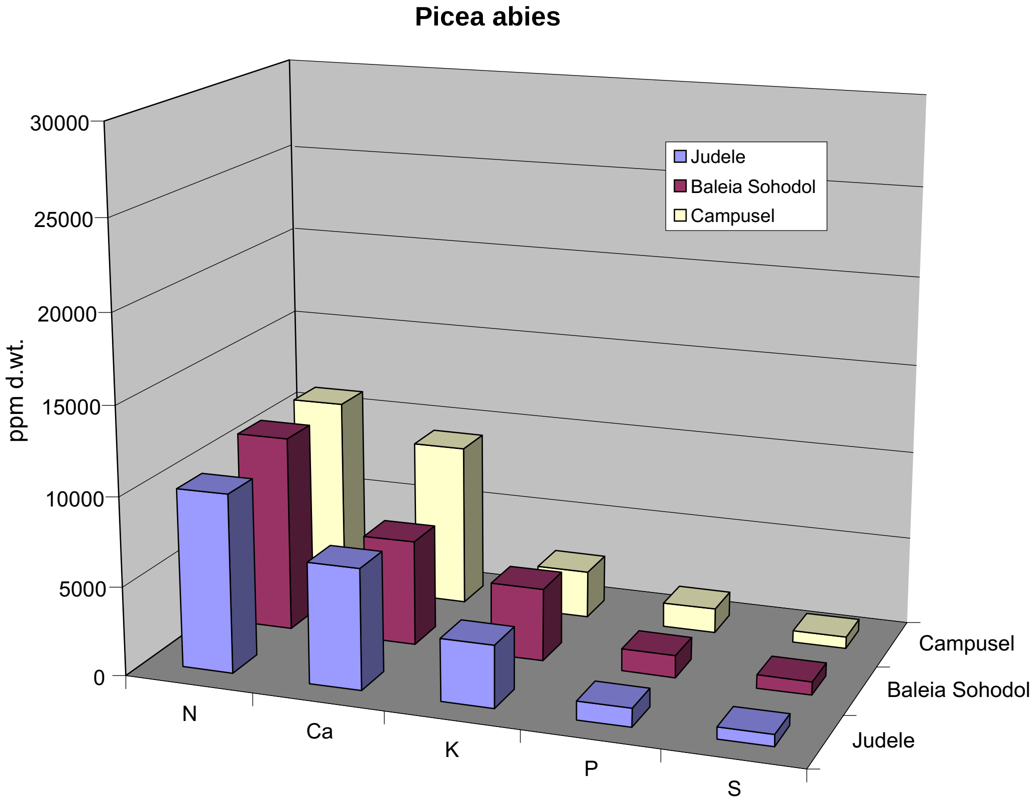


Fig. 11. Concentrations of major nutrients in foliage of *Picea abies*. Two-way ANOVA indicated no significant differences between the study sites for individual nutrients ($F = 10.25 < F_{5\%} = 19.42$).

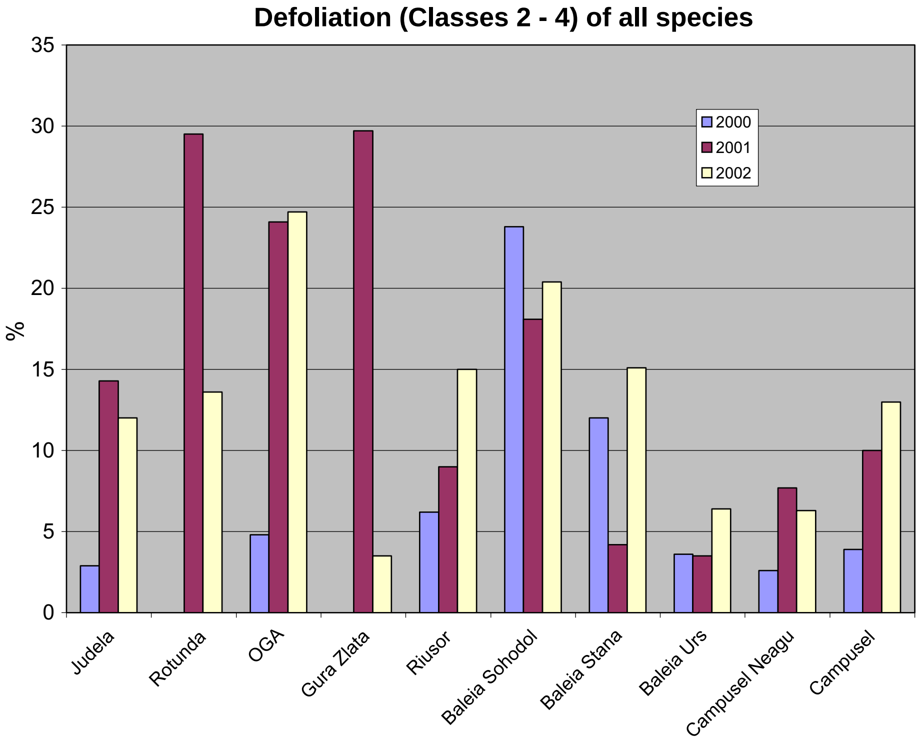


Fig. 12. Percentage of tress with defoliation >25% (classes 2–4) for all tree species in all study sites. Two-way ANOVA indicated no significant effects of year ($F_1 = 0.97 < F_{5\%} = 2.86$) and study site ($F_2 = 5.03 < F_{5\%} = 19.46$).

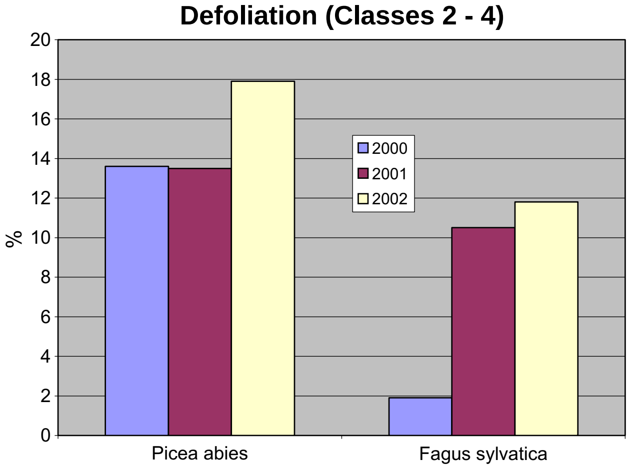


Fig. 13. Defoliation of Norway spruce (*Picea abies*) and European beech (*Fagus sylvatica*) as averages from all sites. No statistical tests could be performed due to the small number of samples.

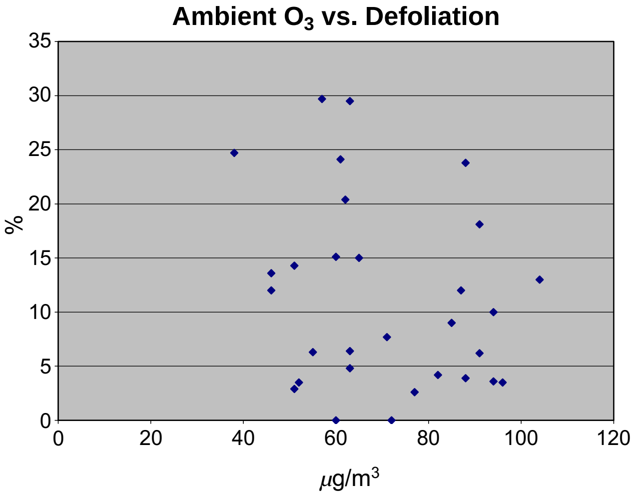


Fig. 14. Relationship between ambient seasonal O_3 concentrations and occurrence of defoliation >25% (classes 2–4) for all tree species during the 2000–2002 period. R^2 for linear relationship was 0.0524 indicating no significant effects of ambient ozone on defoliation.

Table 5
Types of plant communities in research network in Retezat National Park

Site	Plant community
Judele	*Hieracio* (*transsilvanico*), *Picetum* Pawl. Et Br. B1.39, em. Bohr 75, *oxalidentosum*
Rotunda	*Pulmonario* (*rubrae*), *Abieti*, *Fagetum* Soó 64
OGA	*Piceto fagetum abieti* with *Calamagrostis*, *Luzula*
Gura Zlata	*Luzulo fagetum* (Beldie 51) Mor. et al. 68
Riusor	*Piceto fagetum carpaticum* Klika 27 (*Dacicum* Beldie 67, tip *Luzula*, *Clamagrostis*)
Baleia Sohodol	*Hieracio* (*transsilvanico*), *Picetum* Pawl. Et Br. B1.39, em. Bohr 75, *oxalidentosum*
Baleia Stana	*Phyllitidi*, *Fagetum* Vida 1963 (*Acereto*, *Fraxinetum* Pauca 41)
Baleia Urs	*Luzulo fagetum* (Beldie 51) Mor. et al. 68 *myrtilletosum* Soó 62
Campusel Neagu	*Fagetum carpatica* Klika 27, *oxalidentosum* Vida 59
Campusel	*Luzulo fagetum* (Beldie 51) Mor. et al. 68 *myrtilletosum* Soó 62

Table 6
Variation of diameter at breast height (DBH) and height of trees depending on stands type

Stand type	Diameter at breast height (cm)		Height of trees (m)	
	Average	Maximum	Average	Maximum
Spruce-stand	21–30	57–86	17–25	21–34
Beech-stand	22–27	69–124	14–27	18–39
Mixed-stand	16–40	43–106	18–30	24–40

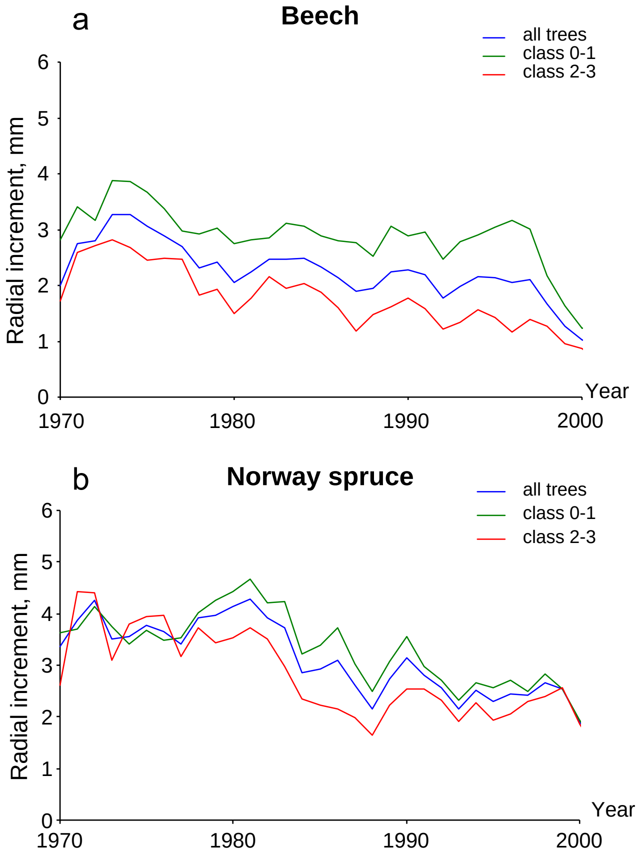


Fig. 15. (a) Annual radial growth of European beech for all trees selected by defoliation group classes 0–1 and 2–3 at the Riusor site. One-way ANOVA showed that growth for trees in 0–1 defoliation classes was significantly different from growth of trees in defoliation classes 2–3 ($p = 5\%$, $F_1 = 22.18 > F_{5\%} = 4.00$). Values of standard deviation for average width of annual rings were $SD_{0-1} = 0.34$ and $SD_{2-3} = 0.49$ mm. (b) Annual radial growth of Norway spruce for all trees selected on defoliation group classes 0–1 and 2–3 at the Riusor site. One-way ANOVA showed that growth for trees in 0–1 defoliation classes was significantly different from growth of trees in defoliation classes 2–3 ($p = 5\%$, $F_1 = 4.43 > F_{5\%} = 4.00$). Values of standard deviation for average width of annual rings were $SD_{0-1} = 1.14$ and $SD_{2-3} = 1.17$ mm.

5. Conclusions

1. In general, the Retezat Mountains experienced good air quality during the three years of this study.
2. Ambient O_3 showed consistent spatial and temporal characteristics: (a) the western portion of the mountains experienced the lowest levels; (b) in 2000 and 2001 concentrations were similar throughout the season, while in 2002 concentrations after mid-August were very low; (c) seasonal average O_3 concentrations in 2002 were much lower than in 2000 and 2001 seasons. Ambient O_3 was not related to the crown condition of European beech and Norway spruce.
3. Wet precipitation, throughfall and soil solutions were highly acidic in most of the studies sites.
4. Crown defoliation was generally low compared with other European forest locations, however, deterioration of crown condition in time was observed. This was probably caused by drought and highly acidic precipitation.
5. Trees with higher defoliation grew less as indicated by annual growth increments.

Acknowledgements

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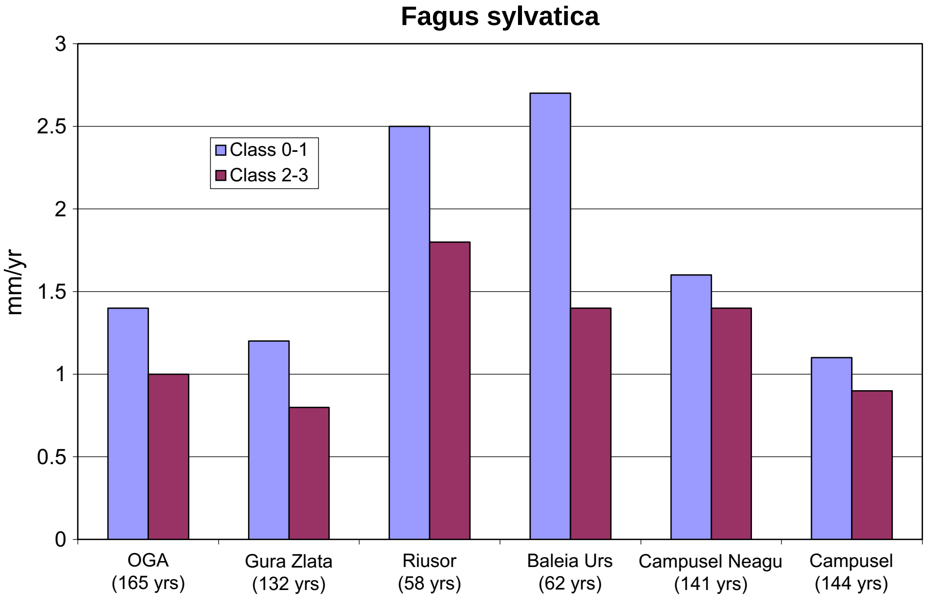

Fig. 16. Average annual radial growth of European beech by defoliation classes (0–1 and 2–3) in individual study sites. Two-way ANOVA showed that average values of annual growth were not significantly different between defoliation group classes 0–1 and 2–3 ($F=2.74<F_{5\%}=243$) and significantly different among study sites ($F=6.02>F_{5\%}=4.70$).

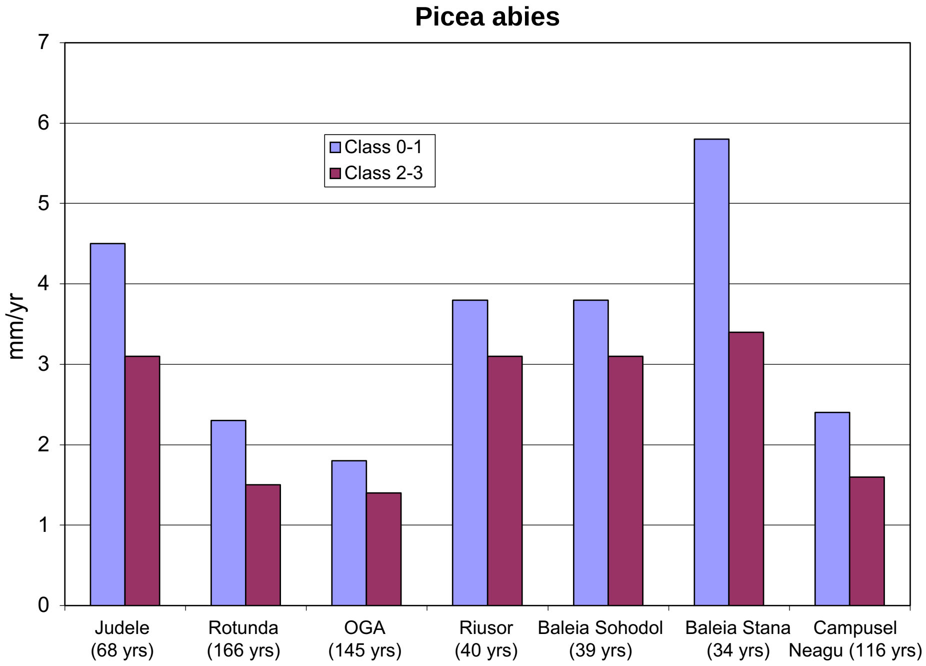

Fig. 17. Average annual radial growth of Norway spruce for defoliation group classes (0–1 and 2–3) in individual study sites. Two-way ANOVA showed that average values of annual radius growth were not significantly different between defoliation group classes 0–1 and 2–3 ($F=2.44<F_{5\%}=245$) and significantly different among study sites ($F=11.37>F_{5\%}=3.56$).

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