Atmospheric Deposition and Ozone Levels in Swiss Forests: Are Critical Values Exceeded?

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Abstract Air pollution affects forest health through atmospheric deposition of acidic and nitrogen compounds and elevated levels of tropospheric ozone (O_3). In 1985, a monitoring network was established across Europe and various research efforts have since been undertaken to define critical values. We measured atmospheric deposition of acidity and nitrogen as well as ambient levels of O_3 on 12, 13, and 14 plots, respectively, in the framework of the Swiss Long-Term Forest Ecosystem Research (LWF) in the period from 1995 to 2002. We estimated the critical loads of acidity and of nitrogen, using the steady state mass balance approach, and calculated the critical O_3 levels using the AOT40 approach. The deposition of acidity exceeded the critical loads on 2 plots and almost reached them on 4 plots. The median of the measured molar ratio of base nutrient cations to total dissolved aluminium (Bc/Al) in the soil solution was higher than the critical value of 1 for all depths, and also at the plots with an exceedance of the critical load of acidity. For nitrogen, critical loads were exceeded on 8 plots and deposition likely represents a long-term ecological risk on 3 to 10 plots. For O_3, exceedance of critical levels was recorded on 12 plots, and led to the development of typical O_3-induced visible injury on trees and shrubs, but not for all plots due to (1) the site specific composition of O_3 sensitive and tolerant plant species, and (2) the influence of microclimatic site conditions on the stomatal behaviour, i.e., O_3 uptake.

Keywords acid deposition · critical levels · critical loads · forest · ICP-Forests Level II · LWF · nitrogen · risk assessment · tropospheric ozone · Switzerland

1 Introduction

The release of pollutants such as sulphur dioxide (SO_2), nitrogen oxides (NO_x) and ammonia (NH_3) into the atmosphere has increased massively during the last century, raising concerns about the effects of resulting changes in atmospheric deposition and ambient air concentrations on forest ecosystems in Central Europe and elsewhere (Percy and Ferretti 2004).

In Switzerland, the concentrations of SO_2, NO_x and NH_3 increased significantly between 1950 and 1990. Since the 1990s, SO_2 concentrations have been reduced to pre-1950 levels through the implementation of various measures. Although NO_x and NH_3 were also reduced, the target concentrations (pre-1950s levels) have not yet been reached (SAEFL 2002).

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Increases in acidic atmospheric deposition, i.e., the deposition of sulphur and nitrogen compounds, have been shown to accelerate soil acidification, possibly leading to toxic levels of aluminium or manganese in the plant-available soil water. The molar ratio between the exchangeable base nutrient cations Ca^{++}, Mg^{++} and K^+ (Bc) and aluminium (Al), Bc/Al in the soil solution, has been widely accepted as an indicator for plant toxicity (Sverdrup and Warfvinge 1993).

Furthermore, nitrogen can act as a fertilizer (Spiecker 1999a). In the case of nitrogen saturation (Aber et al. 1989), increased nitrogen concentrations in soil solution may also lead to nutrient imbalances for trees or to changes in the composition of ground vegetation and increased nitrate leaching into ground or surface waters.

NO_x and other gases are precursors for tropospheric ozone (O_3). Uptake of O_3 at ambient concentrations through the stomata and the related accumulation of reactive oxygen species within plant leaves can cause the collapse of parenchym cells leading to decreased rates of photosynthesis and plant growth (Kärenlampi and Skärby 1996). Ozone-induced injuries present a wide range of visible symptoms (e.g., Gravano et al. 2004; Innes et al. 1996, 2001; Novak et al. 2003).

In the framework of the Convention on Long-Range Transboundary Air Pollution (LRTAP), the International Co-operative Program on the Assessment and Monitoring of Air Pollution Effects on Forests in Europe (ICP-Forests) was established in 1985 (de Vries et al. 2003; Fischer et al. 2005). The ICP-Forests program consists of (1) an annual survey of forest condition on several thousands of Level I plots, and (2) intensive monitoring of forest ecosystems including measurements of atmospheric deposition and air pollutants on about 800 Level II plots. In this context, the concept of critical loads and critical levels (CL) was introduced for the assessment of the effects of air pollution on ecosystems. The CL was defined at the Workshop in Skokloster (Nilsson and Grennfelt 1988) as "a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified elements of the environment do not occur according to present knowledge."

For forest ecosystems, critical loads for acidity (CL_A) (Workshops in Skokloster 1988 and in Copenhagen 1999; see Nilsson and Grennfelt 1988), for nitrogen (CL_N) (Workshops in Skokloster 1988, in Lökeberg 1992, in Geneva 1995, and in Berne 2002; see Achermann and Bobbink 2003; Grennfelt and Thörnelöf 1992) and critical levels of ambient O_3 (CL_{O3}) (Workshops in Kuopio 1996, in Gerzensee 1999, and in Gothenburg 2002; see Kärenlampi and Skärby 1996; Karlsson et al. 2003) were determined or discussed based on the database available at that time, e.g., based on the ICP-Forests monitoring program. Maps of exceedances of critical values were refined using modelled N and S deposition and O_3 concentrations (Posch et al. 2003). However, there is little literature comparing exceedance with the response of forest ecosystems (e.g., Reynolds et al. 1998). Furthermore, monitoring investigations within the ICP-Forests framework were conducted based on the critical loads and levels as defined in the past. In the meantime, however, the broad database of ICP-Forests as well as new findings have initiated further discussions about the air pollutant effects on forest ecosystems and the Level II approach (e.g., Graf Pannatier et al. 2004; Novak et al. 2003; Schaub et al. 2003; Schmitt et al. 2005; Suutari et al. 2001; Thimonier et al. 2005).

Thus, our objectives are (1) to assess the ecological risks, and (2) to evaluate the current CL concept based on the findings of the Swiss Long-Term Forest Ecosystem Research Programme (LWF), the Swiss contribution to the ICP-Forests Level II assessment.

In order to achieve these objectives, we estimated the exceedance of critical loads and levels on the Swiss Level II plots, and we compared indicators for possible effects with the respective levels of exceedance.

2 Methods

2.1 Study sites

The investigations on the Swiss Level II plots were carried out according to the guidelines as described within the ICP-Forests Manual (ICP-Forests 1998 and updates). The plots were selected according to various criteria including the relevance of the forest community type, the homogeneity, and the sensitivity to environmental changes (Cherubini and Innes 2000; Kräuchi 1996; Thimonier et al. 2001) and are located across the five main geographic regions of Switzerland (Fig. 1). Table 1 describes the main characteristics of the plots.

2.2 Sampling and measurements

Each of the 16 Swiss Level II plots covers an area of 1–2 ha. This area includes a 43×43 m intensive monitoring plot, subdivided into 16 subplots of 100 m^2 with a spacing of 1 m between neighbouring subplots. Most measurements for this study were carried out on these subplots. On 13 Level II plots, we determined the solute fluxes in throughfall precipitation in the forest, as well as the fluxes in the bulk precipitation in a nearby open field, which was at a distance between 0.15 and 2.8 km from the respective forested stand. Sampling was carried out with snow (1–4 replicates) or rainfall (3–16 replicates) collectors and bi-weekly collection intervals as described in detail by Thimonier et al. (2005) for at least 1 year during the period from 1995 to 2001 for each plot. The total deposition of sulphur (S), nitrogen (N), and the base cations Ca^{++}, Mg^{++}, K$^+$, and Na$^+$ (BC) was calculated annually based on the canopy budget model of Ulrich (1983), applying the equations recommended in the ICP-Forests Technical Report (de Vries et al. 2001) that include a ratio of 1:6 for the uptake of NH$_4^+$ and H$^+$ by the canopy. On 16 Level II plots, layer-wise sampling and chemical analyses of the soil matrix (16 replications) were carried out during the period from 1995 to 1998 (Walthert et al. 2003). On 8 Level II plots, the soil solution was sampled at depths of 15, 50 and 80 cm every 2 weeks with suction cups (8 replicates) from 1999 to 2002, and analysed chemically as described by Graf Pannatier et al. (2004). The soil solutions was sampled throughout the year except at Celerina, where it was frozen during winter.

On 14 Level II plots, bi-weekly mean ozone concentrations (ppb) were measured throughout the 2002 growing season with passive samplers (Passam, Switzerland). These passive samplers were installed in triplicate at 2 m above ground level in the open-field next to the bulk deposition collectors (ICP-Forests Manual 1998). The measurement, data and quality assurance methods used in connection with these samplers are reported by Sanz, Calatayud, and Sanchez-Peña (2006).

In the same year, an assessment survey of symptoms of O$_3$-induced visible injury was carried out at the light-exposed forest edge closest to the O$_3$ monitoring site.

2.3 Exceedances

We applied the steady-state mass balance approach for calculating critical loads for acidity and nitrogen (Sverdrup and de Vries 1994) and the AOT40 approach for critical ozone levels and their exceedance (Fuhrer et al. 1997), both as described by the Mapping Manual of LRTAP (Spranger et al. 2004).

Fig. 1 Location of the Swiss Long-Term Forest Ecosystem Research (LWF)-plots (ICP-Forests Level II plots of Switzerland)
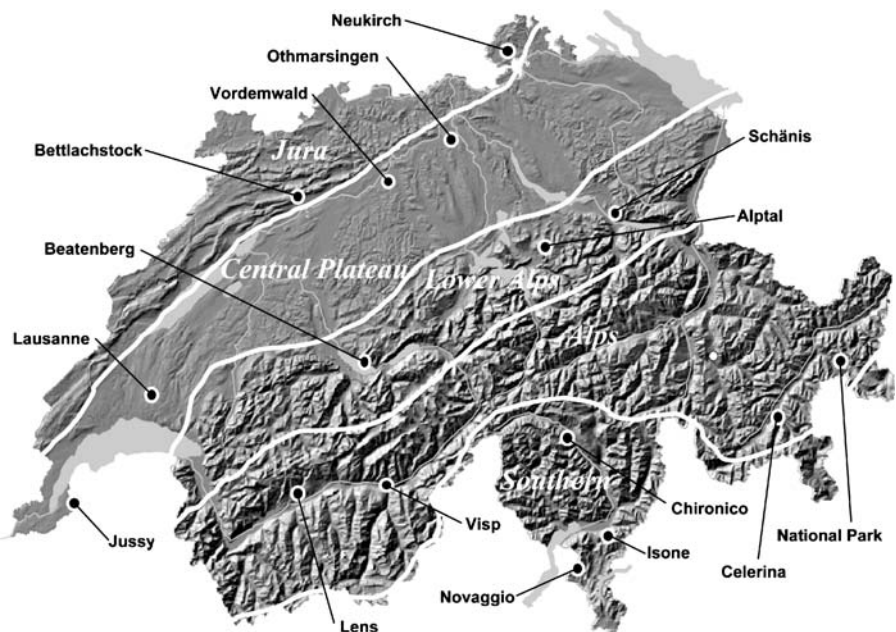
Table 1 Site characteristics of the Swiss long-term forest ecosystem research (LWF)-plots (ICP-forests level II plots of Switzerland)

Region	Jura		Central Plateau				Lower Alps			Alps				Southern Alps		
Plot name	Bettlach-stock	Neun-kirch	Jussy	Lausanne	Othmar-singen	Vordem-wald	Alptal	Beaten-berg	Schänis	Celerina	National Park	Lens	Visp	Chiro-nico	Isone	Novag-gio
Dominant tree species	Fagus sylv.	Fagus sylv.	Quercus robur	Fagus sylv.	Fagus sylv.	Abies alba	Picea abies	Picea abies	Fagus sylv.	Pinus cembra	Pinus mugo	Pinus sylv.	Pinus sylv.	Picea abies	Fagus sylv.	Quercus cerris
Soil type (FAO 1997)	Rendzic Leptosol	Rendzic Leptosol	Eutric Gleysol	Dystric Cambi-sol	Haplic Acrisol	Dystric Planosol	Mollic Gleysol	Podsol	Eutric Cambi-sol	Podsol	Calcaric Fluvisol	Haplic Calcisol	Calcaric Phaeo-zeme	Podsol	Podsol	Podsol
Altitude [m a.s.l.]	1200	600	500	800	500	480	1150	1500	700	1850	1900	1050	700	1350	1200	950
Weathering class rate [eq m^{-3} a^{-1}]	High[a]	High[a]	0.2	0.1	0.1	0.1	0.2	0.05	0.1	0.05	High[a]	N.d.	High[a]	0.05	0.05	0.05
Depth of rooting zone [m]	1.2	1.2	1.2	1.2	1.2	1.2	0.4	0.6	1.2	1.2	1.2	1.2	1.1	1.2	1.2	1.2
Wood harvesting [m^3 ha^{-1} a^{-1}]	4.6	4.6	6.8	6.8	6.8	8.5	4.3	4.3	6.8	2.9	2.9	N.d.	2.8	1.8	0.8	0.8

[a] The soil is well buffered by calcareous contents that dominate its composition.

N.d.: no data available.

2.3.1 Acidity

The exceedance of the critical loads for acidity ExCL$_A$ (keq ha^{-1} a^{-1}) was calculated with

$$\text{ExCL}_A = L_A - CL_A \tag{1}$$

where L$_A$ (keq ha^{-1} a^{-1}) is the acid deposition and CL$_A$ (keq ha^{-1} a^{-1}) the critical load of acidity. L$_A$ was calculated with the following equation:

$$L_A = TD_s + TD_N \tag{2}$$

where TD$_S$ and TD$_N$ are the total atmospheric deposition of sulphur (S) and nitrogen (N), and we used a factor of 2 and 1 eq mol^{-1} of deposited S and N, respectively. The critical loads for acidity CL$_A$ were estimated based on the following equation:

$$CL_A = TD_{BC} + BC_w - Bc_{up}^{net} + N_{up}^{net} + N_i$$
$$+ N_{de} - ANC_{le}^{crit} \tag{3}$$

where TD$_{BC}$ (keq ha^{-1} a^{-1}) is the total atmospheric deposition of base cations (BC = Ca^{2+}, Mg^{2+}, K$^+$ and Na$^+$), BC$_w$ (keq ha^{-1} a^{-1}) is the neutralization capacity produced by the weathering of base cations, Bc$_{up}^{net}$ (keq ha^{-1} a^{-1}) and N$_{up}^{net}$ (keq ha^{-1} a^{-1}) are the net uptake of base cations (Bc = Ca^{2+}, Mg^{2+}, K$^+$) and nitrogen in the tree biomass that is removed permanently from the system by wood harvesting, N$_i$ (keq ha^{-1} a^{-1}) the nitrogen immobilization rate in the soil, N$_{de}$ (keq ha^{-1} a^{-1}) the denitrification rate, and ANC$_{le}^{crit}$ (keq ha^{-1} a^{-1}) the critical leaching of acid neutralising capacity. Bc$_{up}^{net}$ was estimated by multiplying the regional mean wood harvesting volume with the mean wood density, the mean concentration of the base cations in the wood, and their charge (eq mol^{-1}). N$_{up}^{net}$ was estimated similarly using a charge of 1 eq mol^{-1} for N. The nitrogen immobilization rates N$_i$ were approximated according to Rihm's (1994) assignment of N$_i$ values to the soil types of the Swiss soil map. The denitrification was assumed to be neglegible, N$_{de}$ = 0. Two criteria were been applied to define ANC$_{le}^{crit}$: (1) over long term, the ratio of Bc/Al in the soil solution should remain higher than 1 (Bc/Al>1) and (2) Al leaching (Al$_{le}^{3+}$) should not be in excess of the Al produced by the weathering (Al$_w^{3+}$) of primary minerals (Al$_{le}^{3+}$ < Al$_w^{3+}$). The latter criteria aims at preventing the depletion of secondary Al phases and complexes which may cause structural changes in the soil.

It is assumed that an excess of acidic inputs results in leaching of H^+ and Al^{3+} (H_{le}^+, Al_{le}^{3+}) from the soil, according to equation (4):

$$ANC_{le} = H_{le}^+ + Al_{le}^{3+}. \qquad (4)$$

The gibbsite equilibrium equation (5) describes the relationship between the concentrations of H^+ and Al^{3+} in the weathering process, with

$$[H^+] = \frac{[Al^{3+}]}{K_{Gibb}^{1/3}} \qquad (5)$$

where K_{Gibb} is the gibbsite equilibrium constant.

For the conversion of concentrations into fluxes, we estimated the leaching water flux Q (mm a^{-1}) with

$$Q = P - ET - SR \qquad (6)$$

where P (mm a^{-1}) is the measured precipitation, ET (mm a^{-1}) the evapotranspiration, and SR (mm a^{-1}) the surface runoff estimated with

$$ET = a - H b \qquad (7)$$

where H (m) is the altitude of the plot a = 693 mm a^{-1}, b = 0.18 mm $a^{-1}m^{-1}$ and

$$SR = 0.1 \cdot P. \qquad (8)$$

For BC_W, we used Rihm's (1994) estimates of specific weathering rate BC_w^{class} (eq m^{-3} a^{-1}) for each class of the Swiss soil map (Table 1) including his correction for the soil temperature T (K), hence

$$BC_W = BC_w^{class} \cdot D^{-C_1 \left(\frac{1}{T} - \frac{1}{C_2} \right)} \cdot C_3 \qquad (9)$$

where D (m) is the depth of the rooting zone, T (K) the mean annual temperature C_1 = 3800 K, C_2 = 283 K constants and C_3 = 104 m^2 ha^{-1} a unity conversion factor. In an initial run (I), we estimated $D_{(I)}$ was 0.5 m for altitudes below 1600 m a.s.l. and 0.3 m for altitudes above 1600 m a.s.l using the method of Rihm (1994). In a second run (II), we estimated $D_{(II)}$ as the depth to the uppermost limit for roots observed in the soil profile at the plot, such as a permanently oxygen-free horizon, a compact rock surface or a horizon with a fine material density higher than 1.8 g cm^{-3}. We used $D_{(II)}$ = 1.2 m in case that no such limit was found to the depth of 1.2 m.

The first criterion (Bc/Al > 1) as described above was applied by estimating Al_{le}^{3+} with Bc/Al from the leaching of base cations (Bc_{le}), with

$$Al_{le}^{3+} = \frac{1}{Bc/Al} Bc_{le,} \qquad (10)$$

leading to the first equation (11) for Al_{le}^{3+} limitation:

$$Al_{le}^{3+} < Bc_{le} \qquad (11)$$

For Bc_{le} we used

$$Bc_{le} = X_{Bc/BC} BC_W + Bc_{dep} - Bc_{up}^{net}$$
$$- Bc_{le(min)}Q \qquad (12)$$

where $X_{Bc/BC}$ is between 0.6 and 0.8 (eq eq^{-1}) and is the fraction of Ca^{++}, Mg^{++}, and K^+ of the base cations Ca^{++}, Mg^{++}, K^+ and Na^+ supplied by weathering. $Bc_{le(min)}$, the residual Bc concentration in the soil solution below which the trees are not able to take up base cations, was approximated as 15 eq m^{-3}.

The second criterion ($Al_{le}^{3+} \leq Al_w^{3+}$) was applied by using an approximate value of 1.5 for the Al_w^{3+}/Bc_W ratio, which typically ranges from 1 to 3:

$$Al_w^{3+} = 1.5 Bc_W, \qquad (13)$$

leading to the second equation (14) for Al_{le}^{3+} limitation:

$$Al_{le}^{3+} \leq 2.5 Bc_W. \qquad (14)$$

The lower limit for Al_{le}^{3+} from the equations 11 and 14 was used to calculate ANC_{le}^{crit} and the critical load of acidity (CL_A).

2.3.2 Nitrogen

An empirical value for critical loads for nitrogen (CL_N^{emp}) in temperate forests, between 10 and 20 kg ha^{-1} a^{-1}, depending on plot characteristics, was determined at the Workshop in Berne in 2002 (Achermann and Bobbink 2003), and is currently recommended by the LRTAP.

Applying (i) the N saturation and (ii) the steady-state mass balance (SMB) concepts, we calculated the critical loads (CL_N^{SMB}) by taking some of the plot characteristics into account:

$$CL_N^{SMB} = N_{le(acc)} + N_u + N_i + N_{de} \qquad (15)$$

where $N_{le(acc)}$ of 4 and 5 kg ha^{-1} a^{-1} is the acceptable leaching of nitrogen compounds into the surface and subsurface waters for coniferous and deciduous forests, respectively. Here, the denitrification rate N_{de} is approximated with the approach proposed by de Vries at the Workshop in Lökeberg (Grennfelt and Thörnelöf 1992),

$$N_{de} = f_{de}(N_{dep} - N_u - N_i) \qquad (16)$$

applying Rihm's (1996) estimation of f_{de}-values for the drainage condition classes used by the Swiss National Forest Inventory. These f_{de}-values are comparable to the values proposed by Downing et al. (1993).

2.3.3 Ozone

The critical level for O_3 was defined according to the AOT40 concept (Accumulated O_3 over the Threshold of 40 ppb). The AOT40 value is calculated as the sum of the hourly ozone concentrations, $[O_3]_i$ (ppb), exceeding the threshold $C_4 = 40$ ppb over the course of the vegetation period as defined from April 1 to September 30. Only daylight hours when global radiation (R) exceeds $R_{min} = 50$ W m^{-2} are considered (Fuhrer and Achermann 1999; Kärenlampi and Skärby 1996):

$$AOT40 = \sum_{[O_3]_i > C_4\, R\, > R_{min}} ([O_3]_i - C_4) \qquad (17)$$

The critical AOT40 level for ozone (CL$_{O3}$) is based on the criteria to protect the trees from a 10% biomass loss and has been determined to be 5 ppm h (Workshop in Gothenburg 2002). Hourly ozone data $[O_3]_i$ were derived from the measured bi-weekly values based on the function of Loibl and Smidt (1996) which describes the ozone daily profile as a function of relative altitude. The detailed modelling methods for the hourly ozone concentrations are described by Gerosa et al. (2006).

2.4 Effects

2.4.1 Acidification of the soil solution

The Bc/Al ratio (mol mol^{-1}) in the soil solution, where Al stands for the total dissolved aluminium

concentration, was calculated for the 8 Level II plots where the soil solution was sampled. For plots without soil solution sampling, the Bc/Al ratio was approximated from the base saturation BS in the soil matrix with

$$Bc/A1_{le} = m \cdot BS \qquad (18)$$

where m = 0.3, i.e., the lower limit of the range of regression coefficients (0.3–0.5) for the plots where soil solution and soil matrix have both been analyzed (Graf Pannatier et al. 2004).

In addition, the Bc/Al^{3+} ratio, where only the Al^{3+} species is considered, was calculated for 5 Level II plots with acid mineral horizons (Graf Pannatier et al. 2004). We calculated the median of the Bc/Al and Bc/Al^{3+} ratio for each soil depth (15, 50 and 80 cm) and used the lowest of these three ratios as an indicator for the soil acidification status.

2.4.2 Ozone injury

Ozone-induced visible injury was reported in late summer 2002 within the framework of the ICP-Forests program on the Assessment of Ozone Injury on European Forest Ecosystems at the light-exposed forest edge of 16 Level II plots. Symptoms like ozone-induced visible injury observed on native tree and shrub species are validated against symptoms reproduced in open-top chamber facilities, such as in southern Switzerland at the Lattecaldo Cantonal Forest Nursery (e.g., Innes et al. 1996; Innes et al. 2001; Novak et al. 2003) and compared to the extensive ICP-Forests database for ozone injury (see http://www.gva.es/ceam/ICP-forests and http://www.ozone.wsl.ch).

3 Results

For acidity, the atmospheric deposition was calculated for 12 plots and ranged from 0.63 to 3.29 keq ha^{-1} a^{-1} (Fig. 2). The lowest values resulted from low S and N inputs and were measured in the Alps at the plots of Celerina (0.64 keq ha^{-1} a^{-1}) and the National Park (0.63 keq ha^{-1} a^{-1}). For Visp and Lens, no deposition measurements were available. In the Lower Alps, on the Central Plateau and in the Jura, the values ranged from 1.02 to 1.96 keq ha^{-1} a^{-1}, except for Schänis, where the acid deposition was as high as 2.56 keq
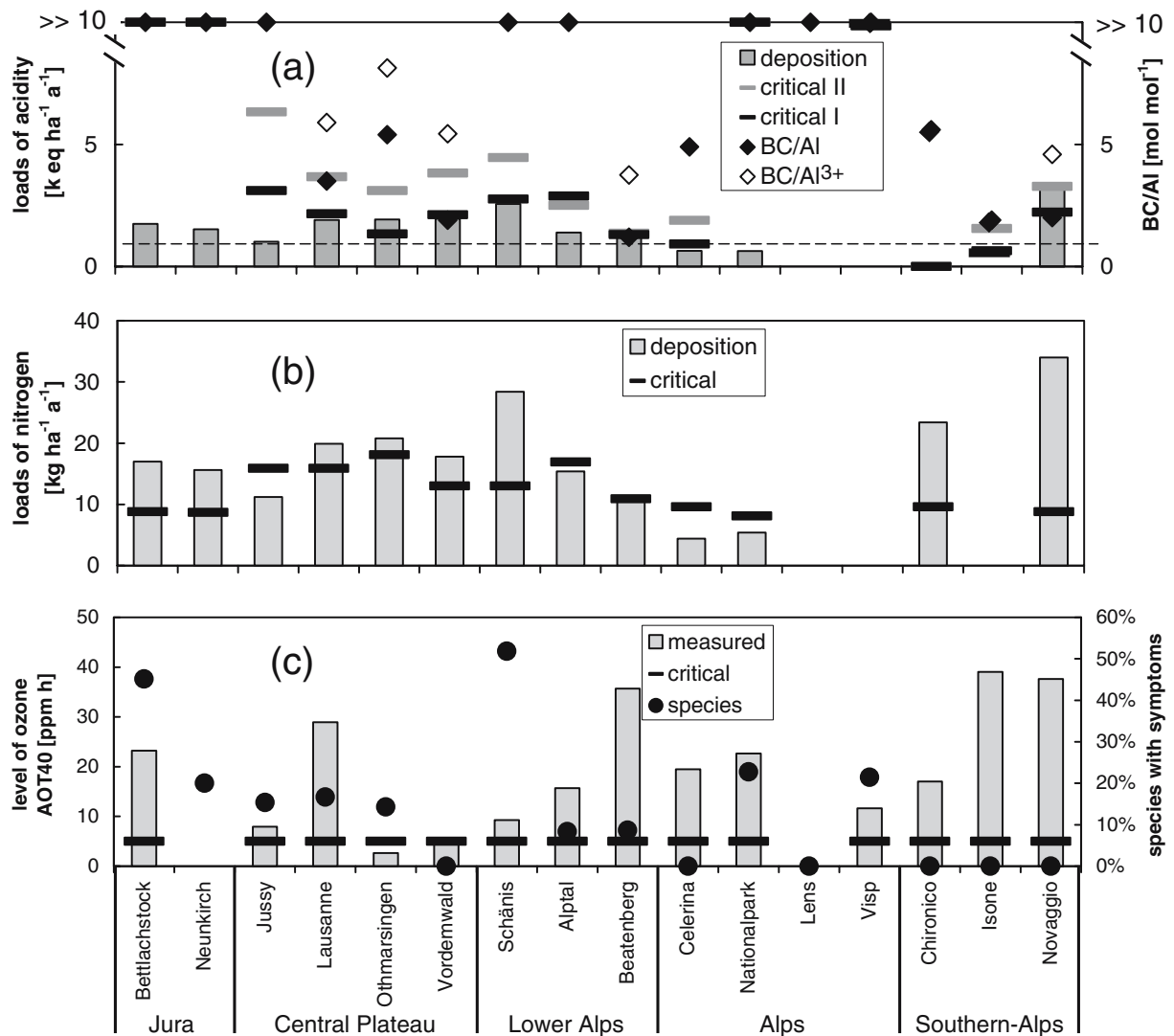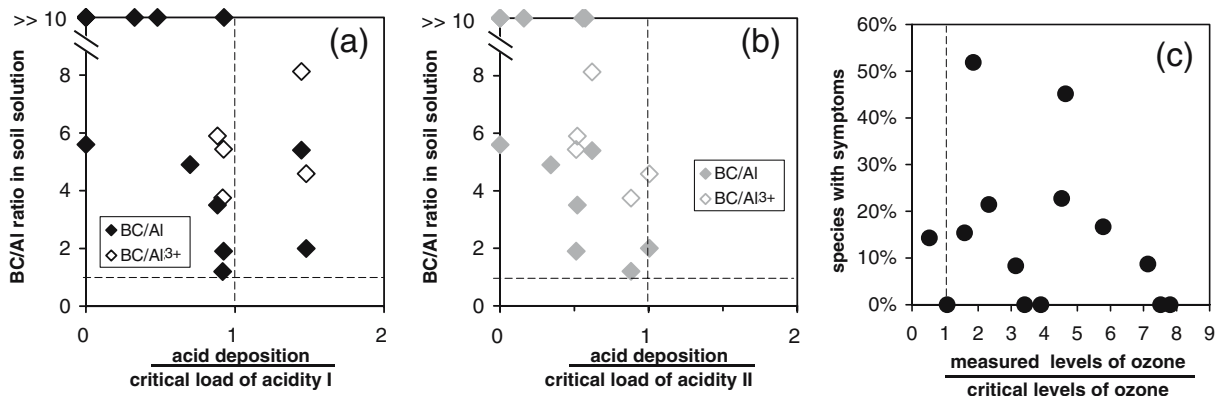
Fig. 2 (a) Atmospheric deposition and critical loads for acidity and the lowest of the median Bc/Al and Bc/Al^{3+} ratio in soil solution at 15, 50, or 80 cm depth. Critical loads were calculated with Rihm's (1994) estimate of the rooting zone depth (critical I) and with a rooting zone depth estimated based on soil profile examinations (critical II). (b) Atmospheric deposition and critical loads for nitrogen. (c) Measured and critical levels of ozone and percentage of species at a nearby forest edge with ozone-induced visible injury symptoms (2002) for ICP-Forests Level II plots of Switzerland

ha^{-1} a^{-1}. The highest deposition was found at Novaggio in the Southern Alps. For Chironico and Isone, no values were available. The critical loads for acidity were very high at the plots in the Jura, at the National Park, and at Visp due to calcareous soils which buffer the acidic inputs. For the other plots, the critical loads for acidity were low. In general the rooting zone depths D$_{(II)}$ estimated based on the soil profile examinations (Table 1) were higher than Rihm's (1994) estimate D$_{(I)}$ (equation 9). Consequently the critical loads for the first run (I) were in general

lower than those for the second run (II), except for Alptal (I: 2.9 keq ha^{-1} a^{-1} II: 2.5) and Beatenberg (I: 1.3, II: 1.4). The acid deposition exceeded the critical loads of the first run (I) at 2 plots (at Novaggio by 1.0 keq ha^{-1} a^{-1} and at Othmarsingen by 0.6) and approached them at 4 plots (Lausannne, Vordemwald, Schänis, Beatenberg) but they did not exceed the critical loads of the second run (II) (Fig. 2).

The lowest Bc/Al ratios in the soil profile (Fig. 2) were close to the critical value of 1 and were measured at Beatenberg (1.2), Vordemwald (1.9),

Fig. 3 (a) Lowest of the median Bc/Al and Bc/Al^{3+} ratio in soil solution at 15, 50 or 80 cm depth plotted against the ratio of atmospheric deposition to critical loads for acidity I calculated in the initial run (I) with root zone depths according to Rihm (1994). (b) Lowest of the median Bc/Al and Bc/Al^{3+} ratio in soil solution at 15, 50 or 80 cm depth plotted against the ratio of atmospheric deposition to critical loads for acidity II calculated in the second run (II) with rooting zone depth estimated based on the soil profile examinations. Bc/Al < 1 indicates aluminium toxicity. (c) Percentage of species at nearby forest edge with ozone-induced visible injury symptoms vs. ratio of actual ozone level over critical ozone level

and Novaggio (2.0), where the acid deposition approached the critical loads. In contrast, the Bc/Al ratios were much higher at Othmarsingen (5.4), Lausanne (3.5) and Schänis (>10) although that the acid deposition approached or even exceeded the critical loads at these plots (see also Fig. 3a and b).

For N, deposition was available for 13 plots. It ranged from 4.4 to 34 kg N ha^{-1} a^{-1} and its pattern was similar to that for acid deposition. The critical loads for nitrogen (CL_N^{SMB}) were within the range from 8 to 20 kg N ha^{-1} a^{-1} and thus less variable than those for acidity. They were exceeded at Novaggio (25 kg N ha^{-1} a^{-1}) and Chironico (14) in the Southern Alps, at Schänis (15) in the Lower Alps, and at 5 plots on the Central Plateau and in the Jura. Further, they were approached at the other 2 plots in the Lower Alps (Alptal and Beatenberg). In the Alps (Celerina and the National Park), the deposition of nitrogen was far below the critical load.

For ozone, actual AOT40 levels ranged from 2.6 to 39 ppm h in 2002 (Fig. 2) and its spatial pattern differed from that of the deposition. The highest levels were measured at Isone (39.0 ppm h) and Novaggio (37.6) in the Southern Alps, at Beatenberg (35.7) in the Lower Alps and at Lausanne (28.9) on the Central Plateau. The lowest levels were registered at Othmarsingen (2.6) and Vordemwald (5.3) on the Central Plateau. The critical AOT40 level of 5 ppm h was exceeded on 12 out of 14 plots.

Figure 3c shows the percentage of the species which were growing at the forest edge closest to the plot and developed ozone-induced visible injury. Highest percentages of affected species were found at Schänis (52%), Bettlachstock (45%), National Park (23%), and Visp (21%). Despite high exceedances of the critical level, few symptoms of ozone-induced visible injury were found at Beatenberg (9%), Alptal (8%), and Lausanne (17%) and no ozone-induced visible injury was found at Novaggio, Isone, and Celerina (Fig. 3c). Conversely, we found typical ozone-induced visible injury at Schänis (52%) and Othmarsingen (14%) although the critical AOT40 level was not exceeded there.

4 Discussion

The relatively high deposition loads of S and N at Novaggio and Schänis are typical for areas at the edge of sharply rising mountain chains in the proximity of industrialized and densely populated areas such as the Italian Po Plain and the Swiss Central Plateau (Della Lucia et al. 1996; Schmitt et al. 2005; Thimonier et al. 2005). These exposed areas are subjected to similar high pollutant concentrations in the air as in the plain, but receive higher precipitation. In contrast, Celerina and National Park are located within mountain chains, remote from densely populated areas.

Since around 1985, the loads of acidity have decreased in Europe, mainly due to a reduction in sulphur dioxide emissions from road traffic and industrial activities (SAEFL 2002). Klöti et al. 1989

determined a sulphur deposition of about 1.8 keq ha^{-1} a^{-1} at Lägern in the Central Plateau of Switzerland between June 1986 and June 1987, while our annual values for S deposition between 1995 and 2001 ranged from 0.1 to 1.0 keq ha^{-1} a^{-1}. In comparison, in 1988 during the massive forest decline, Dambrine et al. (1993) measured S and N deposition of 4 and 3 keq ha^{-1} a^{-1}, respectively, at Naèetin in the Krušné hory Mts. (Erzgebirge, Czech Republik) where the soils are poorly buffered. According to Kurz et al. (1998), the critical loads for acidity were exceeded for about 60% of the forested area in Switzerland using the 1986–1990 acid deposition. They determined median acid deposition of about 3.5, 4.5 and 2.5 k eq ha^{-1} a^{-1} for the Jura, the Central Plateau, and the Alps, respectively. Hence, the reduction of sulphur emission successfully decreased the acid deposition to below the critical loads for acidity for numerous plots (see also Posch et al. 1999).

However, the critical loads for acidity presented here include uncertainties such as the estimate of the weathering rate as shown by varying the main rooting zone depths (D), applied in equation (9). Our impression that Rihm's (1994) estimate of D seems to underestimate the rooting zone depths remains to be proven. Moreover, the uncertainty of the specific weathering rate could be reduced substantially by including the soil chemistry and mineralogy measurements into the calculations of the weathering rates and by using dynamic models. Such a soil chemistry model (PROFILE) was applied by Kurz et al. (1998) for 770 sites in Switzerland with a known chemistry of the soil matrix, but the model has neither been calibrated nor validated with Swiss soil solution chemistry measurements.

No relationship could be found between the exceedance of critical loads and the Bc/Al ratios. The field data indicate that the ecological risks related to the critical Bc/Al ratio might be lower than those estimated with the model. When determining the speciation for Al in the soil solution, Graf Pannatier et al. (2004) found that much of the Al consists of less toxic Al species rather than Al^{3+} at Beatenberg and Vordemwald. At Vordemwald, Othmarsingen, and Lausanne, the lowest Bc/Al ratios were found in the uppermost layer at 15 cm depth with increasing values towards the lower layers (Graf Pannatier 2004). Adapted root systems may compensate for the low ratio in the topsoil by increased uptake of base

cations from lower layers (Walthert et al. 2006). In addition, Grigal and Ohmann (2005) have shown that upwards diffusion of cations may also be important and should be considered for the CL models. In contrast, at Beatenberg, the lowest Bc/Al ratio was recorded at 80 cm depth in the mineral layer clearly below the rooting zone and may thus not affect the plant roots (Graf Pannatier et al. 2004).

However, the concept is based on the steady-state assumption, although the atmospheric deposition has changed in the past few decades. Therefore, the Bc/Al ratio is not necessarily expected to correlate to the exceedance at the moment, but over long-term. As the soil solution sampling on the Swiss Level II plots started only in 1999, no trend analysis could be conducted for Bc/Al ratios. Consequently, we have no information whether the measured state is the result of soil formation or it is, in addition, influenced by accelerated soil acidification during the last few decades. Long-term measurements of the Bc/Al ratio at Copera in Southern Switzerland showed a decreasing trend during the last 16 years (Graf Pannatier et al. 2005). Hence, despite the successful reduction of the sulphur emission, a long-term risk of an accelerated soil acidification is still likely for some forested areas in Switzerland with poorly buffered soils and high atmospheric deposition of acidity, e.g., at Novaggio. One possible effect, a lower storm resistance of trees, has been recently discussed by Mayer et al. (2005) and others.

The atmospheric deposition of nitrogen still exceeded the critical loads for a number of plots, in particular for two plots in the Southern Alps (Novaggio: TD$_N$-CL$_N^{SMB}$ = 25.2 kg N ha^{-1} a^{-1}, Chironico: 13.8) and for one plot in the Lower Alps (Schänis: 15.4). The high N deposition is largely related to the emissions from road traffic, agriculture, industry, and heating systems in Switzerland and the neighbouring countries. Most of the calculated CL$_N^{SMB}$ were within the range of CL$_N^{EMP}$. The SMB calculations of the critical loads for nitrogen include uncertainties, especially the terms of N$_{de}$ and N$_i$. The uncertainties might be in the range of the difference between calculated deposition and critical loads for up to about 7 plots. Further, the SMB concept does not take into account the actual N saturation status of a plot. At Alptal for example, experimentally added N was still immobilized in the soil (Schleppi et al. 1998) and we suppose that many other sites are not yet N saturated.

Increasing soil N pools can be remobilized after disturbances such as clear-cuts or storm damage (Hegg et al. 2004). Consequently, the atmospheric deposition of nitrogen may represent a long-term risk for forest ecosystems at several sites in Switzerland leading to increasing growth rates, nutrient imbalances, or reduced stress tolerance for trees and increased nitrate leaching (see, e.g., Flückiger and Braun 1999; Mayer et al. 2005; Spiecker 1999b).

The levels of ozone on 14 Swiss Level II plots were spatially variable and the AOT40 values ranged from 2.6 to 39 ppm h. The high levels at Isone (39.0 ppm h), Novaggio (37.6), Beatenberg (35.7), Lausanne (28.9) and Bettlachstock (23.2) might be related to (1) the proximity of these plots to emission sources such as intensively used traffic axes and industrialized agglomerations, and (2) to the exposure of the emitted precursors to sunlight and high temperatures. Slightly or considerably lower levels were registered at plots which are very remote from larger cities such as the National Park (22.6), Celerina (19.5), and Alptal (15.7). Yet, lowest levels were recorded at plots in close vicinity of NO_x emission sources such as Othmarsingen (2.6) and Vordemwald (5.3). These low levels can be explained by the fact that high NO_x levels enhance ozone decomposition.

The levels of ozone exceeded the critical AOT40 value at 12 out of 14 plots. The derivation of AOT40 values from bi-weekly passive sampling measurements implies some uncertainty. Furthermore, ozone concentrations are subject to high inter-annual variability and we included measurements from 2002 only. The species composition at the forest edges and their sensitivity to ozone differ from plot to plot. This limits the comparability of the quota of plant species showing ozone-induced visible injury versus O_3-tolerant species.

On the other hand, plants may respond to increased vapour pressure deficit and reduced soil water availability by reducing the stomatal opening, leading to a reduced O_3 uptake despite peak ozone levels (e.g., Schaub et al. 2003). This influence by environmental site conditions is not taken into account with the AOT40 concept as stated for example by G. P. Karlsson, P. E. Karlsson, Soja, Vanermeiren, and Pleijel (2004). In the Southern Alps, for example, in particular at Novaggio and Isone, high summer temperatures combined with low relative humidity leading to high vapour pressure deficits at the leaf level limit the stomatal uptake. These conditions are common and frequent for southern Switzerland, which may explain the lack of ozone-induced visible injury despite of the occurrence of some of the highest O_3 concentration values across Europe. In order to take the influence of environmental factors on the physiological plant behaviour into account, the estimate of a critical O_3 uptake may be more biologically relevant than the concentration based AOT40 concept (e.g., Matyssek et al. 2004).

5 Conclusions

The atmospheric deposition of acidity exceeded the critical loads at 2 plots and approached them at 4 out of 12 Swiss Level II plots. Therefore, we cannot exclude long-term ecological risks related to Al toxicity and Al depletion in Switzerland in areas with high deposition loads of acidity and poorly-buffered soils. The results of the critical loads calculations with the steady-state mass balance showed no relationship with the measured Bc/Al ratio in the soil solution, but we cannot exclude a correlation in future when the system reaches steady state. However, the frequently used method that we applied here includes a large uncertainty due to estimation method of the weathering rate. The calculation of critical loads could be substantially improved by (1) modelling the weathering process based on the physical and chemical measurements on the plots, (2) using dynamic models, (3) validating the models with the measured soil solution chemistry, (4) including Al speciation, and (5) cation diffusion. In addition, long-term soil solution chemistry data are of key importance for the assessment of the soil acidification risk in Swiss forests.

For nitrogen, the critical loads were clearly exceeded for 3 out of 5 Swiss Level II plots in the Lower and the Southern Alps, whereas they were not exceeded at the plots in sparsely populated areas in the Alps. In the Jura and on the Central Plateau, the critical loads were lower but were still exceeded at 5 out of 6 plots. Considering calculation uncertainties, the actual deposition is likely to represent a long-term risk for 3 to 10 out of 13 plots. In the long-term, such exceedance may lead to increasing tree growth, decreased stress tolerance of trees, nutrient imbalan-

ces, changes in species composition, and increased nitrate leaching into ground water.

Critical levels of O₃ were exceeded on 12 out of 14 Swiss Level II plots. The quota of the plant species showing ozone-induced visible injury does not correspond to the exceedance of the AOT40 critical level of 5 ppm h O₃. Partly, this lack of correlation may be due to the differing species composition of O₃-sensitive versus O₃-tolerant species. Another source of uncertainty may lie in the estimation of the AOT40 statistics based on data from bi-weekly passive sampling. The variation due to the differing species composition among plots may be eliminated with the planting of O₃-sensitive bio-indicator species, which would allow a sound comparison of onset and severity of negative ozone effects such as visible injury among the plots. The calculation of the effective ozone uptake rather than the exceedance of a certain concentration-based threshold, in combination with the bio-indicator approach would, furthermore, allow a more biologically meaningful ozone risk assessment for forest ecosystems.

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