



Nitrogen can act as a fertilizer and enhance forest growth (Spiecker, 1999). However, elevated nitrogen inputs represent a potential threat to the stability of forest ecosystems: enhanced nitrogen deposition can lead to nutrient imbalances, soil acidification, changes in the composition of the ground vegetation, changes in mycorrhizae, and increased susceptibility to secondary stresses (Skeffington and Wilson, 1988). “Nitrogen saturation” may be reached when the availability of inorganic nitrogen is in excess of the plant and microbial nutritional demand and the ecosystem is unable to retain all the incoming nitrogen (Aber *et al.*, 1989), leading to increased nitrogen leaching below the rooting zone. The sensitivity of forest ecosystems to nitrogen deposition in the long term has been formalized using the concept of “critical loads”, defined as a quantitative estimate of an exposure to a nitrogen deposition below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge (Nilsson and Grennfelt, 1988).

Two approaches can be used to quantify atmospheric deposition on a given location: (i) measurements on the site itself, or (ii) application to the site of a model calibrated for the region of interest using available datasets, surveys and field information.

In Switzerland, the first approach (measurements) has been used since the mid-1980s to quantify wet and dry deposition at a few forest sites in projects which were often motivated by a growing concern over the effects of air pollution on forest health (e.g. Flückiger *et al.*, 1986; Kloeti *et al.*, 1989; Haemmerli *et al.*, 1992; Flückiger and Braun, 1998; Schleppi *et al.*, 1998). In the mid-1990s, a network of 17 selected forest sites was set up within the framework of the Swiss Long-Term Forest Ecosystem Research (LWF, according to the German: Langfristige Waldökosystem-Forschung) to improve our understanding of how natural and anthropogenic stresses affect forests in the long term (Cherubini and Innes, 2000; Thimonier *et al.*, 2001). LWF meets the aims of the International Co-operative Programme on the Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests), which was implemented under the Convention on Long-range Transboundary Air Pollution of the UN/ECE (EC-UN/ECE *et al.*, 2001).

On selected LWF plots, atmospheric deposition is assessed with the throughfall method (Draaijers *et al.*, 1996). This method relies on the parallel sampling of precipitation under the forest canopy (throughfall) and precipitation in a nearby open area. Throughfall reflects the composition of both incident precipitation (wet deposition) and dry deposition, as the substances deposited during dry periods on branches and foliage is partly washed out during subsequent precipitation. Wet deposition is obtained directly from sampling in the open field if “wet-only” collectors (which are open only when it rains) are used. If continuously open collectors are used (bulk precipitation), correction is needed for deposition of particulate material on the collectors during dry periods. Dry deposition on forests is considerably higher than dry deposition in the open field due to the roughness of the canopy, the large surface areas developed by the foliage and the physical

80 and physiological properties of the leaves (cuticular adsorption, stomatal uptake of  
81 gases). Estimates of dry deposition on forests can be derived from the difference  
82 between throughfall and wet or bulk precipitation, so long as a number of assump-  
83 tions about the canopy exchange processes (uptake and leaching of nutrients) are  
84 made.

85 The second approach to quantifying nitrogen deposition (modeling) involves  
86 first calculating concentrations in the atmosphere. This can be done either by us-  
87 ing emission inventories and chemical transformations of each emitted component  
88 in simulating atmospheric dispersion (transport model), or by extrapolating point  
89 measurements using known geographical parameters such as land use, topography  
90 or road networks (geo-statistical methods). The wet deposition is then obtained  
91 by multiplying the concentrations in precipitation with the precipitation amounts,  
92 and the dry deposition by multiplying air concentrations with deposition velocities  
93 (inferential modeling). These deposition velocities depend on the reactivity of the  
94 compound, land use, vegetation type and, for some models, on additional factors  
95 such as boundary layer turbulence. The deposition of sulfur and nitrogen across  
96 Europe has been calculated with this approach at a spatial resolution of  $50 \text{ km} \times$   
97  $50 \text{ km}$  and a high temporal resolution (EMEP, see e.g. Jonson *et al.*, 1998). The  
98 approach was adapted in the mid-1990s by Rihm (1996) so that the regional distri-  
99 bution of nitrogen deposition in Switzerland could be calculated at a higher spatial  
100 resolution ( $1 \text{ km} \times 1 \text{ km}$ ).

101 In this paper we present the results of throughfall sampling on 13 LWF sites, for  
102 which up to 7 years of measurements are available. We also analyze the output of  
103 the deposition model developed by Rihm (1996), which was run for these 13 sites  
104 to calculate recent annual wet and dry deposition of nitrogen and sulfur. We address  
105 then three main questions: (1) How well does the atmospheric deposition derived  
106 from throughfall measurements compare with the modeled nitrogen and sulfur  
107 deposition? (2) Where do the inconsistencies, if any, come from? (3) On which  
108 LWF plots does nitrogen deposition exceed the critical load and thus represent a  
109 potential risk for the forest ecosystem?

110

## 2. Methods

### 111 2.1. STUDY SITES

112 Throughfall chemistry was monitored on the 13 LWF plots selected for at least  
113 one year. Precipitation was sampled in parallel using continuously open collec-  
114 tors (bulk precipitation) in open areas close to the forest plots. The collectors  
115 in the open area were next to a meteorological station also run by LWF that  
116 recorded precipitation with a tipping bucket, air temperature, humidity, solar ra-  
117 diation, UV-B and PAR radiation, and wind speed and direction. The study sites  
118 are located in various regions of Switzerland. Two plots (Bettlachstock (BET),



Figure 1. Location of LWF plots (results for the plot in Visp are not given as sampling only started in 2001) (Swiss Federal Statistical Office/Federal Office of Topography).

Neunkirch (NEU)) are located in the Jura mountains; four (Jussy (JUS), Lausanne 119 (LAU), Othmarsingen (OTH), Vorderwald (VOR)) on the Central Plateau; three 120 (Alptal (ALP), Beatenberg (BEA), Schanis (SCH)) in the Lower Alps; two 121 (Celerina (CEL), National Park (NAT)) in the Alps; and two (Chironico (CHI), Novag- 122 gio (NOV)) in the Southern Alps (Figure 1). The site characteristics are shown in 123 Table I. 124

## 2.2. SAMPLING DESIGN IN THE FIELD 125

Throughfall collectors were divided into two sets of equal numbers, and systemati- 126 cally distributed in two 43 m × 43 m subplots inside the forest plot. One of each pair 127 of subplots is dedicated to the intensive monitoring of soil chemistry (solid phase 128 and soil solution), litterfall and ground vegetation in addition to throughfall. The 129 other subplot, which is adjacent to the intensive monitoring subplot in most cases, 130 was designed to cover the spatial variability of the forest stand better. Collectors of 131 the same design were used in the forest stands and the open area, but the type and 132 number of collectors installed varied depending on the amount of snowfall expected 133 during winter. 134

(a) *Sampling design when no significant snowfall was expected (funnel type col- 135 lectors):* Funnel-type collectors were used throughout the year on plots where 136 snow precipitation is not expected to be abundant in winter (NEU; JUS, LAU, 137

TABLE I  
 Site description. Jussy is divided in two subplots, one unmanaged, one managed. Long-term mean annual precipitation amounts were derived from precipitation maps established for the period 1972–1992 (FOWG, 2000)

Region	Site	Longitude	Latitude	Altitude a.s.l. (m)	Main tree species	Age of the main tree species (yr)	Mean precipitation (mm)	Sampling years
Jura	BET	07°25	47°14	1149	<i>Fagus sylvatica</i>	170	1549	1999–2001
	NEU	08°32	47°41	582	<i>Fagus sylvatica</i>	150	1044	1998
Central Plateau	JUS	06°17	46°14	501	<i>Quercus robur</i>	80	977	1998–2001
	LAU	06°40	46°35	807	<i>Fagus sylvatica</i>	150–160	1210	1997–2001
Lower Alps	OTH	08°14	47°24	484	<i>Fagus sylvatica</i>	110–130	1123	1995–2001
	VOR	07°53	47°17	480	<i>Abies alba</i>	100	1115	1997–2001
Alps	ALP	08°43	47°03	1160	<i>Picea abies</i>	170–220	2286	1997–1998
	BEA	07°46	46°43	1511	<i>Picea abies</i>	180–220	1305	2000–2001
Alps	SCH	09°04	47°10	733	<i>Fagus sylvatica</i>	120–140	1801	1999–2001
	CEL	09°53	46°30	1871	<i>Pinus cembra</i>	200–240	1024	1999–2001
Southern Alps	NAT	10°14	46°40	1899	<i>Pinus mugo</i>	200	1048	1999–2001
	CHI	08°49	46°27	1365	<i>Picea abies</i>	150–170	2187	2001
	NOV	08°50	46°01	950	<i>Quercus cerris</i>	60	2017	1997–2001

OTH, VOR) and only during the snow-free season on plots where heavy snowfall is likely in winter (BET; ALP, BEA, SCH; CEL, NAT; CHI, NOV) (Figure 1).

The funnel-type collectors consisted of a funnel with a 100 cm<sup>2</sup> opening, connected to a 2 l storage bottle (see also description in Thimonier (1998a, 1998b)). The collectors were installed in PVC-pipes in order to shield the samples from direct solar radiation, and were equipped with guard rings to reduce the risk of contamination by bird droppings. Contamination with organic material was minimized by using two filters. The first was a polyethylene disk (diameter 55 mm) placed inside the funnel to prevent contamination with coarse material and the second a polyester net fitted between the funnel and the storage bottle to avoid contamination with fine material. Initially, this net consisted of a piece of fabric with a mesh size of 30  $\mu$ m. From March 2001 onwards, however, we used a net with a larger mesh size (250  $\mu$ m) and a tighter connection between the funnel and the storage bottle to prevent water running on the outer wall flowing into the bottle through this net (Draaijers *et al.*, 2001).

Bulk precipitation was sampled with three collectors in the open area. The opening of the collector was set at a height of 1.5 m above ground. In the forest stand, throughfall was sampled either with 16 collectors or, at CHI, with 8 collectors at a height of 1 m and distributed over the two 43 m  $\times$  43 m subplots. At OTH, where throughfall sampling began before the sampling design was standardized for all plots, the collectors were originally located in lines (Thimonier, 1998b). The intensive monitoring subplot on which eight collectors were subsequently relocated was installed in 1998. We kept only the eight collectors, which were in use throughout the whole 1995–2001 period for our calculations.

Precipitation samples were collected every two weeks and either directly brought back to the laboratory or sent by post in insulated boxes. On each sampling occasion, the whole collector (funnel, filters and storage bottle) was replaced by a clean one that had been acid washed or rinsed with deionized water in the laboratory.

*(b) Sampling design when abundant snowfall was expected (bucket-type collectors or plastic bags):* Snow collectors were used in winter on plots where snow precipitation is expected to be abundant. Two types of collectors were used: 30-cm diameter buckets equipped with plastic bags before and after the winter period 1998–1999 and long plastic bags fitted to a 20 cm diameter PVC ring during the winter 1998–1999. This second type of snow collector with a smaller surface area was selected to reduce the frequency of the bucket collectors overflowing, which occurred on a number of occasions during the previous winter. However, the smaller areas made the collecting less efficient as, under certain conditions, a snowcap formed, so buckets were used again in the following winters.

Access to the plots is difficult in winter, as the roads are often closed due to snow, so that it is necessary to walk further carrying a lot of equipment and samples with greater risks of avalanche danger. Hence, the number of collectors was significantly reduced in winter, with only one collector in the open area, and four in the forest

180 stand (except at CHI where no snow sampling was performed in the stand). The  
181 four snow collectors in the forest stand were distributed on the two 43 m × 43 m  
182 subplots and each was placed close to a funnel-type collector location.

183 Precipitation samples were collected every two weeks, except at BET and CEL  
184 where the collection interval was extended to four weeks for logistic reasons.  
185 A new plastic bag was installed in the buckets (or the PVC rings). When sam-  
186 pling was carried out by the WSL staff, the whole sample was taken back to  
187 the laboratory. When sampling was carried out by a local forester, the amount  
188 of precipitation was determined with a spring balance and a subsample of the  
189 melted sample was sent by post in insulated boxes to the laboratory. On a few  
190 occasions when the collectors were filled with snow and access to the plot was  
191 particularly difficult, all four collectors were weighed but only a subset was car-  
192 ried back for chemical analyses. The remaining collectors were emptied in the  
193 field.

### 194 2.3. SAMPLE PREPARATION AND CHEMICAL ANALYSES

195 The time between sample collection and sample preparation was generally shorter  
196 than 3 days, and sometimes samples could be prepared within a day. All sam-  
197 ples were either prepared immediately upon arrival or stored at 2 °C. During its  
198 preparation, each sample was weighed and its conductivity measured (reference  
199 temperature 20 °C). If bird droppings in the funnels or the buckets were reported on  
200 the field form, the sample was discarded. In addition, conductivity was used to detect  
201 possible contamination of the bulk precipitation samples. The spatial variability of  
202 atmospheric deposition should be rather low in open areas. Under this assumption,  
203 where funnel-type collectors were used, one of the three samples was discarded  
204 if it showed a conductivity that exceeded the average of the conductivities of the  
205 other two by more than 2  $\mu\text{S cm}^{-1}$  or by more than 10%. The remaining samples  
206 were then merged, resulting in composite samples of either bulk precipitation or  
207 throughfall.

208 For each sample its pH was measured and it was then filtered (0.45  $\mu\text{m}$ ) and pre-  
209 pared in duplicate, one for chemical analysis, the other for storage at either -20 °C  
210 samples for analyzing ammonium) or +2 °C (other samples) until validation of the  
211 analyses. Ammonium ( $\text{NH}_4^+$ ) was determined colorimetrically with automated flow  
212 injection analysis (FIA). Calcium (Ca), magnesium (Mg), potassium (K), sodium  
213 (Na) and total phosphorus (P) were determined on acidified samples (1.13%  $\text{HNO}_3$ -  
214 Suprapur) with inductively coupled plasma-mass spectrometry (ICP-MS). From  
215 March 2000 onwards, these elements were determined with inductively coupled  
216 plasma-atomic emission spectrometry (ICP-AES). Nitrate ( $\text{NO}_3^-$ ), sulfate ( $\text{SO}_4^{2-}$ )  
217 and chloride ( $\text{Cl}^-$ ) were identified using ion chromatography (IC). The total dis-  
218 solved nitrogen (TDN) was measured with alkaline persulfate oxidation (converting  
219 all nitrogen into nitrate) until the beginning of 2001. After March 2001, TDN was  
220 analyzed using a TOC-V analyser (Shimadzu, Tokyo, Japan) after acidification to

pH 2–3 with combustion at 720 °C (converting all nitrogen to nitrogen monoxide) 221  
 and chemiluminescence gas detection. Our quality controls showed that the alka- 222  
 line persulfate oxidation method led to irregular contaminations of the samples and 223  
 the TOC-V analyser seemed to underestimate TDN. Therefore fluxes of dissolved 224  
 organic nitrogen ( $\text{DON} = \text{TDN} - \text{NH}_4\text{-N} - \text{NO}_3\text{-N}$ ) were only calculated approxi- 225  
 mately and were not included in the atmospheric deposition estimates, as discussed 226  
 later in this paper. All the analyses were checked against certified standards and 227  
 the WSL laboratories regularly participate in international comparison exercises 228  
 (e.g. Mosello *et al.*, 1998). Checking of the analytical results was based on the ion 229  
 balance and comparing the measured and calculated conductivity (Rossum, 1975; 230  
 Mosello *et al.*, 1998). Duplicates of questionable samples were re-analyzed. 231

#### 2.4. ESTIMATING MISSING PRECIPITATION AMOUNTS AND CONCENTRATIONS 232

For 2.7% of all collection intervals the precipitation exceeded the storage capacity of 233  
 the collectors and the precipitation amount had to be estimated. We used regressions 234  
 of precipitation amounts measured by our collectors against the precipitation data 235  
 from the tipping bucket installed in the open area and on the forest plots, or against 236  
 the data of the closest stations of the Swiss Meteorological Institute (MeteoSwiss). 237  
 Missing element concentrations on a given plot were similarly estimated using 238  
 regressions between parameters or between concentrations in bulk and throughfall 239  
 samples observed for that plot. 240

At CHI, where throughfall was measured during summer only, the amount of 241  
 winter throughfall was estimated from the winter bulk precipitation using a linear 242  
 regression of the amount of summer throughfall against the summer bulk pre- 243  
 cipitation. The N and S concentrations of the winter throughfall were set to the 244  
 volume-weighted means of N and S concentrations in the summer throughfall. A 245  
 test of this approach with the data of a comparable plot, BEA, resulted in rela- 246  
 tive differences between reconstructed and measured annual fluxes of  $-4.4\%$  for 247  
 inorganic N and  $+4.9\%$  for S on average over 3 years of measurements. 248

#### 2.5. ESTIMATION OF ATMOSPHERIC DEPOSITION FROM THROUGHFALL 249 MEASUREMENTS 250

Throughfall (TF) and bulk precipitation (BP) fluxes were calculated multiply- 251  
 ing precipitation amount and concentrations for each collection interval and were 252  
 summed over calendar years, using interpolations at transitions between years. To 253  
 estimate the wet (WD) and dry (DD) components of the total atmospheric deposition 254  
 on the forest (TD) from TF and BP, several corrections have to be made. 255

To estimate WD, BP must be corrected for dry deposition onto the funnels. We 256  
 applied average wet-only to bulk ratios ( $f_{\text{corr}}$ ) given in the literature (Draaijers *et al.*, 257

258 1998) to our annual fluxes of bulk precipitation.

$$WD = BP \times f_{\text{corr}} \quad (1)$$

259 To estimate TD, we used the canopy budget model developed by Ulrich (1983),  
260 extended by Draaijers (1995) and synthesized in EC-UN/ECE *et al.* (2001). In this  
261 model throughfall fluxes represent TD plus or minus canopy exchanges (CE), which  
262 are the result of canopy leaching (Cle) and canopy uptake (CU).

$$TF = TD + CE = TD + Cle - CU \quad (2)$$

263 For sodium ( $\text{Na}^+$ ), chloride ( $\text{Cl}^-$ ) and sulfate ( $\text{SO}_4^{2-}$ ), canopy exchange CE is  
264 negligible (e.g. Lindberg and Lovett, 1992) and TD is calculated as:

$$TD = TF \quad (3)$$

265 To estimate the total deposition of base cations,  $\text{Na}^+$  is taken as a tracer having  
266 a TD/BP ratio similar to TD/BP for  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{K}^+$  (particles containing  
267  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{K}^+$  are assumed to have the same mass median diameter as  $\text{Na}^+$   
268 containing particles):

$$TD_{\text{bc}} = TF_{\text{Na}}/BP_{\text{Na}} \times BP_{\text{bc}} \quad (4)$$

269 where bc stands for  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  or  $\text{K}^+$ .

270 Canopy leaching of these cations ( $\text{Cle}_{\text{bc}}$ ) is calculated as:

$$\text{Cle}_{\text{bc}} = TF_{\text{bc}} - TD_{\text{bc}} \quad (5)$$

271 We calculated Equations (1)–(5) on an annual basis. This approach was suc-  
272 cessfully tested against the method using micrometeorological measurements and  
273 inferential modeling (Draaijers and Erisman, 1995).

274 For nitrogen compounds, estimating atmospheric deposition using throughfall  
275 data is more uncertain than for other elements. Several studies showed that nitrogen  
276 in gas form ( $\text{NO}_2$ ,  $\text{HNO}_3$ ,  $\text{NH}_3$ ) or in solution ( $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ) can be taken up by  
277 foliage (see review by Harrison *et al.*, 2000). In the extended canopy budget model  
278 (EC-UN/ECE *et al.*, 2001), canopy uptake (CU) of  $\text{NH}_4^+$  and  $\text{H}^+$  is assumed to be  
279 equal to the total canopy leaching of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{K}^+$  taking place through ion  
280 exchange, corrected for the leaching of weak acids (wa).

$$CU_{\text{NH}_4} = (\text{Cle}_{\text{Ca}} + \text{Cle}_{\text{Mg}} + \text{Cle}_{\text{K}}) - \text{Cle}_{\text{wa}} - CU_{\text{H}} \quad (6)$$

281 with

$$CU_{\text{H}} = 6TF_{\text{H}}/(TF_{\text{NH}_4} + 6TF_{\text{H}})(\text{Cle}_{\text{Ca}} + \text{Cle}_{\text{Mg}} + \text{Cle}_{\text{K}} - \text{Cle}_{\text{wa}}) \quad (7)$$

and 282

$$Cle_{wa} = TF_{wa} - 2BP_{wa} \quad (8)$$

where  $w_a$  was calculated as the difference in equivalent concentration of cations (Ca<sup>2+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>, Na<sup>+</sup>, H<sup>+</sup>, NH<sub>4</sub><sup>+</sup>) minus strong acid anions (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>). 283 284

Assuming that NH<sub>4</sub><sup>+</sup> has an exchange efficiency six times larger than NO<sub>3</sub><sup>-</sup>, 285  
canopy uptake of N is calculated as (in equivalent): 286

$$CU_N = CU_{NH_4}(TF_{NH_4} \cdot 6 + TF_{NO_3})/(TF_{NH_4} \cdot 6) \quad (9)$$

Total deposition of nitrogen is then calculated as: 287

$$TD_N = TF_N + CU_N \quad (10)$$

Dry deposition of all compounds is calculated as: 288

$$DD = TD - WD \quad (11)$$

## 2.6. MODELED DEPOSITION OF NITROGEN AND SULFUR 289

Sulfur and nitrogen deposition on the LWF plots was calculated using the combined 290  
simplified model described by Rihm (1996), Kurz *et al.* (1998) and Rihm and Kurz 291  
(2001). Recent monitoring results for Southern Switzerland (Barbieri and Pozzi, 292  
2001) and from a new NH<sub>3</sub>-monitoring program carried out at 40 stations (Thöni 293  
*et al.*, in press) have been included into the model, together with an update of 294  
the SO<sub>2</sub>-concentration map. The model calculating N deposition at the 1 km × 295  
1 km spatial resolution in Switzerland was parameterized so that the total nitrogen 296  
deposition in Switzerland calculated with this model approximately matched the 297  
deposition calculated by EMEP with a resolution of 50 km × 50 km (77.4 kt N a<sup>-1</sup> in 298  
1998, corresponding to 19 kg N ha<sup>-1</sup> a<sup>-1</sup> on average (EMEP, 2000)). The following 299  
compounds were considered in the calculation of N deposition: NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> for 300  
wet deposition, and nitric acid vapor (HNO<sub>3</sub>), the gases nitrogen dioxide (NO<sub>2</sub>) 301  
and ammonia (NH<sub>3</sub>), and particulate NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> (aerosols) for dry deposition 302  
(Equation (12)). The calculation of S deposition included the calculation of the wet 303  
deposition of SO<sub>4</sub><sup>2-</sup> and the dry deposition of sulfur dioxide (SO<sub>2</sub>) (Equation (13)). 304  
Dry deposition of particulate SO<sub>4</sub><sup>2-</sup>, which amounts to only a small fraction of the 305  
wet deposition (Hertz and Bucher, 1990), was not calculated. 306

$$TD_N = WD_{NH_4} + WD_{NO_3} + DD_{NH_3} + DD_{NO_2} + DD_{HNO_3} \\ + DD_{NH_4} + DD_{NO_3} \quad (12)$$

$$TD_S = WD_{SO_4} + DD_{SO_2} \quad (13)$$

307 (a) *Wet deposition:* Wet deposition was obtained by multiplying the mean annual  
308 concentrations of N and S compounds in precipitation with precipitation amounts  
309 from the mean annual precipitation map established by the Federal Office for Water  
310 and Geology (Table I). This map is based on about 300 precipitation measure-  
311 ment stations (FOWG, 2000).  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations were set to  
312 values derived from 12 monitoring stations and stratified according to altitude.  
313 Mean annual concentrations of  $\text{SO}_4^{2-}$  in precipitation were considered to be ho-  
314 mogenous below 1000 m and to decrease linearly with altitude above this limit  
315 (Table II). For  $\text{NO}_3^-$  and  $\text{NH}_4^+$ , a distinction was made between two regions.  
316 North of the Alps, the mean annual concentrations of N compounds in precipi-  
317 tation were, similar to  $\text{SO}_4^{2-}$ , considered to be homogenous below 1000 m and  
318 to decrease linearly with altitude above this limit. Due to the topography of the  
319 south face of the Alps, where the mountain chain acts as a specific barrier to the  
320 southern air mass transport,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations in precipitation were  
321 calculated as a function of altitude, longitude and latitude, according to the re-  
322 gression model of Barbieri and Pozzi (2001) also described by Barbieri *et al.*  
323 (2004).

324 (b) *Dry deposition:* Dry deposition of aerosols and gases was calculated by mul-  
325 tiplying modeled air concentrations (annual means) with deposition velocities for  
326 forests taken from the literature (Rihm, 1994, 1996). For particulate  $\text{NO}_3^-$  and  $\text{NH}_4^+$ ,  
327 concentrations (derived from a few point measurements) and deposition velocities  
328 were stratified according to altitude. In each class of altitudes, concentrations and  
329 deposition velocities were calculated by linear interpolation of the lower and upper  
330 limits of three altitude ranges (Table II). For  $\text{HNO}_3$ , concentrations were similarly  
331 stratified according to altitudes but the deposition velocity was set to a constant.  
332  $\text{NH}_3$  and  $\text{NO}_2$  concentrations were derived from concentration fields calculated  
333 from emission inventories to which statistical dispersion models were then applied  
334 (SAEFL, in press; Thöni *et al.*, in press). The deposition velocities for  $\text{NO}_2$  and  
335  $\text{NH}_3$  were set to values depending on the type of stand (coniferous or broadleaved).  
336 For  $\text{NH}_3$ , the distinction between coniferous and broadleaved stands was not made  
337 in the calculations in Rihm (1996), and the deposition velocity was then set to a  
338 single value ( $30 \text{ mm s}^{-1}$ ).  $\text{SO}_2$  concentrations were mapped by geo-statistical in-  
339 terpolation of 52 stations and the deposition velocity was independent of the forest  
340 type.

341 The years of reference for  $\text{SO}_4^{2-}$  concentrations in precipitation were 1997–  
342 1999. For concentrations of N compounds in precipitation, the years of reference  
343 were 2000 north of the Alps, and 1993–1998 south of the Alps (Barbieri and Pozzi,  
344 2001). The mean precipitation amounts were based on long-term precipitation data  
345 (1972–1992). For air concentrations, the year of reference was 2000 for all nitrogen  
346 compounds and  $\text{SO}_2$ .

TABLE II

Calculation of wet deposition (a) and dry deposition (b) of N and S on LWF plots with the modeling approach

Compound	Altitude range (m)	Concentration ( $\mu\text{eq l}^{-1}$ )		
		North of the Alps	South of the Alps	
(a) Concentrations of N and S in precipitation;				
Wet deposition				
NH <sub>4</sub>	≤1000	32.1	Function of altitude, longitude, latitude (Barbieri & Pozzi, 2001)	
	2000	24.3		
NO <sub>3</sub>	≤1000	23.6	Function of altitude, longitude, latitude (Barbieri & Pozzi, 2001)	
	2000	17.9		
SO <sub>4</sub>	≤1000	22.0		
	2000	17.5		
		Deposition velocity ( $\text{mm s}^{-1}$ )		
Compound	Altitude range (m)	Concentration ( $\mu\text{g m}^{-3}$ )	Coniferous forests	Broadleaved forests
(b) Air concentrations of N and S and deposition velocities				
Dry deposition				
Aerosols				
NH <sub>4</sub>	400	2.02	2	2
	800	1.69	4	4
	2000	0.43	4	4
NO <sub>3</sub>	400	2.79	2	2
	800	2.36	4	4
	2000	0.56	4	4
SO <sub>4</sub>		Not calculated		
HNO <sub>3</sub>	200	1		
	1000	0.5	15	15
	2000	0.25		
Gases				
NO <sub>2</sub>	Emission inventory and dispersion model		4	3
NH <sub>3</sub>	Emission inventory and dispersion model		30	22
SO <sub>2</sub>	Geo-statistical interpolation		8	8

## 2.7. CRITICAL LOADS FOR NITROGEN

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We compared the deposition data derived from our throughfall measurements and the modeled data with the critical loads for nitrogen ( $CL_N$ ). The values of these critical loads were derived using two approaches: (1) the steady state mass balance

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350

351 method (SMB), based on the nitrogen saturation concept (the nitrogen deposition  
 352 must not lead to a situation where the availability of inorganic nitrogen is in excess  
 353 of the total combined plant and microbial nutritional demand) (UBA, 1996; cited  
 354 in Rihm, 1996, and Rihm and Kurz, 2001); and (2) the empirical method, based on  
 355 data obtained from controlled chamber and field experiments, chemical analyses,  
 356 comparisons of vegetation composition in time and space, ecosystem models and  
 357 biological knowledge (Grennfelt and Thörnölöf, 1992; Rihm, 1996; Rihm and Kurz,  
 358 2001).

359 The SMB method uses the following equation (in Rihm, 1996):

$$CL_N = U_N + I_N + le(acc)_N / (1 - fde) \quad (14)$$

360 where  $U_N$  is the amount of N removed by wood harvesting,  $I_N$  the acceptable  
 361 immobilization rate of N in soil organic matter,  $le(acc)_N$  the acceptable total leaching  
 362 from the rooting zone, and  $fde$  the denitrification fraction.

363 The removal by wood harvesting,  $U_N$ , was calculated by multiplying the N  
 364 content of the stems with the long-term harvesting rate. The N content of stems  
 365 was calculated using a wood density of  $500 \text{ kg m}^{-3}$  and a N content of 0.10%  
 366 for coniferous forests (VOR, ALP, BEA, CEL, NAT, CHI), a wood density of  
 367  $700 \text{ kg m}^{-3}$  and a N content of 0.15% for deciduous forests (BET, NEU, JUS,  
 368 LAU, OTH, SCH, NOV). For the long-term wood harvesting rates, the means  
 369 for the five regions of Switzerland was taken (Central Plateau:  $6.8 \text{ m}^3 \text{ ha}^{-1} \text{ a}^{-1}$   
 370 for the deciduous forests (JUS, LAU, OTH),  $8.5 \text{ m}^3 \text{ ha}^{-1} \text{ a}^{-1}$  for the coniferous  
 371 forests (VOR); Lower Alps:  $4.1 \text{ m}^3 \text{ ha}^{-1} \text{ a}^{-1}$  for the deciduous forests (SCH),  
 372  $4.3 \text{ m}^3 \text{ ha}^{-1} \text{ a}^{-1}$  for the coniferous forests (ALP, BEA); Alps:  $2.9 \text{ m}^3 \text{ ha}^{-1} \text{ a}^{-1}$  for  
 373 coniferous forests (CEL); Southern Alps:  $1.6 \text{ m}^3 \text{ ha}^{-1} \text{ a}^{-1}$  for coniferous forests  
 374 (CHI)). For unmanaged plots  $U_N$  was set to 0.

375 The immobilization rate,  $I_N$ , was set to  $3 \text{ kg N ha}^{-1} \text{ a}^{-1}$  at low altitudes ( $< 500 \text{ m}$ ),  
 376  $5 \text{ kg N ha}^{-1} \text{ a}^{-1}$  at high altitudes ( $> 1500 \text{ m}$ ), and calculated by linear interpolation  
 377 at altitudes in-between.

378 The acceptable total leaching,  $le(acc)_N$ , was set to  $4 \text{ kg N ha}^{-1} \text{ a}^{-1}$  for forests  
 379 in the lowlands below  $500 \text{ m}$  (OTH, VOR) and  $2 \text{ kg N ha}^{-1} \text{ a}^{-1}$  for alpine forests  
 380 above  $2000 \text{ m}$ . It was calculated by linear interpolation at altitudes between  $500$  and  
 381  $2000 \text{ m}$  (all plots except OTH, VOR). These values have been changed compared  
 382 to Rihm (1996), where  $le(acc)_N$  was set to  $4 \text{ kg N ha}^{-1} \text{ a}^{-1}$  for coniferous forests  
 383 and  $5 \text{ kg N ha}^{-1} \text{ a}^{-1}$  for deciduous forests.

384 A value for  $fde$  was ascribed to each plot according to the degree of  
 385 hydromorphy derived from the 1:200,000 soil suitability map of Switzerland  
 386 (Bodeneignungskarte der Schweiz 1:200,000, 1980), as done by Rihm (1996).  
 387 Lower values of  $fde$  were ascribed to dry soils, and higher values to wet  
 388 soils.

389 Empirical critical loads were attributed to each plot according to the forest  
 390 type (UBA, 1996):  $7\text{--}20 \text{ kg N ha}^{-1} \text{ a}^{-1}$  for acidic coniferous forest (VOR, BEA,

CEL, CHI), 10–20 for acidic deciduous forests (JUS, LAU, NOV, OTH; SCH), and 391  
15–20 for calcareous forests (BET, NEU; ALP; NAT). 392

### 3. Results 393

#### 3.1. PRECIPITATION AND CANOPY INTERCEPTION 394

Annual precipitation varied markedly from site to site (Table III). The wettest 395  
sites were SCH, ALP (Lower Alps) and NOV (Southern Alps) with precipitation 396  
amounts in the open often exceeding 2000 mm. The lowest annual precipitations 397  
were recorded in NEU (Jura), CEL, NAT (Alps) and JUS (Plateau). Precipitation 398  
amounts varied considerably between the sampling years. NOV had the largest 399  
difference between years, with the precipitation recorded in 2000, the wettest sam- 400  
pling year (2771 mm in the open area) 68% higher than that measured in 1997, the 401  
driest year (1654 mm). 402

The relative rainfall interception was highest at OTH, JUS (broadleaved stands) 403  
and VOR (mixed stand), and lowest at ALP, NAT, CEL and CHI (coniferous stands), 404  
which are located at moderate and high elevations. The inter-annual variability of 405  
interception was high, probably due to the variable intensity and form (snow or 406  
rain) of the single precipitation events, which contribute to the overall pattern. 407

#### 3.2. CONCENTRATIONS 408

Concentrations in bulk precipitation were on average dominated by  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , 409  
 $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$ . They were highest at NOV and NEU, whereas they were lowest at 410  
the two high-elevation sites (CEL, NAT) (Figure 2, Table III). 411

As a rule, the concentrations in throughfall were much higher than those in 412  
bulk precipitation, due to the enrichment of precipitation as it passes through the 413  
canopy (Figure 2). Concentrations were higher in throughfall than bulk precipita- 414  
tion for all elements (most clearly  $\text{K}^+$  and  $\text{Mg}^{2+}$ ), except for  $\text{H}^+$  and, at CEL and 415  
NAT,  $\text{NH}_4^+$ . On these two plots, the concentrations of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  were lower 416  
in throughfall than in bulk precipitation in some of the sampling years. Lower 417  
 $\text{NH}_4^+$  concentrations in throughfall than in bulk precipitation were also observed 418  
at NOV and ALP in some years. pH in the bulk precipitation and throughfall was 419  
lowest at ALP and NOV. The only plots where  $\text{H}^+$  concentrations were noticeably 420  
higher in throughfall than in bulk precipitation for all sampling years were VOR 421  
and BEA. The ranking of the plots according to the solute concentrations was not 422  
the same for bulk and throughfall precipitation, which again illustrated the effect of 423  
the canopy on precipitation chemistry. Concentrations in throughfall were highest 424  
at the plots located on the Central Plateau (especially OTH), the Jura and the South- 425  
ern Alps (NOV). Concentrations in throughfall were lowest at the high-elevation 426  
sites. 427

TABLE III  
Annual precipitation and volume-weighted mean annual element concentrations (ueq/l), median pH and mean conductivity at 20 °C (uS/cm) in the open field (bulk precipitation BP) and under the canopy (throughfall TF)

Site	Year	mm	[NH <sub>4</sub> <sup>+</sup> ]		[NO <sub>3</sub> <sup>-</sup> ]		[SO <sub>4</sub> <sup>2-</sup> ]		[Ca <sup>2+</sup> ]		[Mg <sup>2+</sup> ]		[K <sup>+</sup> ]		[Na <sup>+</sup> ]		[Cl <sup>-</sup> ]		[H <sup>+</sup> ]		Median pH		Conductivity			
			BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF
Jura																										
BET	99	1627	1469	23.8	30.1	29.6	52.3	22.5	32.0	21.8	51.7	3.6	13.7	4.4	37.7	13.5	18.1	8.6	17.7	11.1	9.3	5.5	5.6	9.6	17.5	
	00	1309	1149	21.6	27.0	23.3	43.9	22.1	32.8	23.5	52.9	3.4	14.1	4.0	39.9	9.1	16.9	9.0	18.2	4.3	4.6	5.5	5.8	9.7	19.7	
	01	1622	1446	18.5	23.4	18.3	37.9	17.8	25.8	20.8	46.4	3.3	11.7	3.2	31.8	6.5	9.8	6.2	11.2	3.3	2.0	5.8	6.4	7.7	13.8	
NEU	98	790	648	46.2	55.8	34.8	63.6	33.7	50.2	21.4	55.3	5.0	19.5	4.8	92.3	14.9	18.5	13.0	27.5	7.1	4.9	5.2	6.1	13.4	32.6	
Central Plateau																										
JUS*	98	883	667	24.3	44.5	26.5	30.0	26.1	36.9	22.9	64.6	3.9	34.6	5.3	119.2	10.6	16.7	9.4	24.3	11.2	2.7	5.0	6.0	12.0	30.2	
	99	1316	1039	28.4	53.2	32.5	41.8	26.2	36.5	32.5	75.5	4.7	29.1	3.1	98.1	15.3	20.8	14.1	23.1	5.4	2.9	5.8	6.2	10.3	26.9	
	00	1042	809	25.0	34.5	27.5	29.2	21.8	31.0	34.9	69.8	5.3	31.0	4.0	81.0	10.7	14.9	13.2	20.0	2.8	1.8	5.7	6.2	10.6	25.2	
LAU	01	1224	946	21.6	39.3	22.3	27.6	22.3	30.3	33.1	59.9	4.4	22.6	3.6	75.7	7.9	10.2	7.2	14.4	2.6	0.7	5.9	6.6	8.9	21.3	
	97	1214	1033	44.0	51.8	33.3	53.8	34.8	46.6	24.5	41.7	3.8	16.4	4.8	71.4	11.3	14.3	10.6	17.1	9.7	4.8	5.1	5.9	13.5	24.6	
	98	1181	970	38.9	55.9	32.9	56.8	31.4	44.0	20.0	46.6	4.4	21.1	3.1	65.6	13.5	17.5	11.3	20.6	9.6	7.0	5.2	5.9	13.5	25.3	
OTH	99	1477	1390	41.2	40.2	34.2	50.5	30.1	35.3	38.1	46.0	4.7	17.3	2.7	75.5	16.8	20.6	12.2	17.4	4.4	4.7	5.7	6.0	12.0	21.3	
	00	1361	1067	35.6	48.1	31.7	48.1	25.4	35.0	27.3	44.3	4.2	15.0	2.9	51.0	9.9	14.6	10.5	17.5	4.0	2.2	5.7	6.3	11.7	21.3	
	01	1536	1297	31.8	35.2	24.3	33.3	22.4	28.4	24.4	39.8	3.5	13.0	3.1	50.0	7.3	10.2	5.9	11.4	2.0	1.4	6.1	6.5	8.7	16.4	
VOR	95	1130	854	35.6	70.3	31.1	59.9	31.7	74.3	15.0	59.7	2.8	26.1	3.5	92.5	7.4	13.1	6.1	17.2	13.3	3.7	5.1	5.9	16.1	36.9	
	96	1004	731	44.3	89.3	37.5	78.7	34.5	86.3	15.5	64.7	2.9	24.4	3.1	96.2	11.0	17.8	8.9	19.0	11.6	4.8	5.0	6.4	15.0	33.3	
	97	870	626	36.4	86.3	29.8	73.1	28.1	73.2	14.9	60.4	3.3	20.8	6.4	91.4	9.7	17.0	9.7	21.2	10.8	2.7	5.2	6.2	11.4	33.5	
VOR	98	1024	741	34.1	93.6	27.8	69.6	28.7	71.0	19.6	57.8	4.8	21.5	9.3	87.7	13.1	20.5	13.6	25.5	12.1	4.4	5.1	6.2	12.6	35.1	
	99	1356	1011	33.1	67.2	33.0	60.2	25.8	56.8	25.1	58.2	5.7	21.4	6.4	92.4	13.8	21.3	11.2	18.2	5.1	2.3	5.8	6.3	10.8	29.1	
	00	1077	776	38.4	77.5	33.5	72.2	28.4	58.8	29.5	61.1	4.1	16.0	3.2	56.8	10.9	17.5	10.6	20.5	3.5	3.6	5.6	6.3	12.0	29.0	
ALP	01	1304	982	28.5	53.8	22.6	44.0	21.4	42.7	21.1	51.2	3.7	14.9	4.3	61.0	7.1	11.3	6.2	13.5	2.5	0.8	5.9	6.4	8.7	21.1	
	97	943	633	47.8	74.2	32.3	71.8	28.7	62.8	14.8	49.4	3.1	17.9	5.5	87.9	11.1	21.0	8.9	31.2	7.3	30.8	5.2	5.2	11.4	40.1	
	98	1234	889	39.3	52.1	26.0	58.1	25.9	49.5	17.2	41.1	3.3	15.6	2.9	56.5	11.9	20.0	10.6	25.4	6.4	12.8	5.2	4.9	11.2	26.4	
Lower Alps	99	1342	1030	44.6	52.5	37.0	69.4	26.5	46.1	28.6	52.8	5.3	15.8	3.4	44.4	15.8	22.8	14.1	24.9	6.0	11.4	5.9	5.4	11.3	24.5	
	00	1113	835	40.7	72.0	29.8	70.8	25.2	44.5	21.7	49.6	3.3	14.1	3.0	39.8	10.0	17.6	11.3	20.2	3.1	4.4	5.8	5.5	10.9	25.2	
	01	1406	987	33.8	43.6	23.8	42.6	20.8	33.5	20.7	44.0	3.4	13.7	3.5	64.9	7.3	13.5	6.9	19.6	1.2	9.1	6.1	5.8	8.5	23.5	
ALP	97	2082	2069	26.3	25.6	21.3	25.8	18.4	19.3	6.4	10.7	1.3	2.7	1.7	11.0	8.1	8.4	5.5	6.0	15.4	10.5	4.8	5.0	9.8	10.2	
	98	2093	2027	19.9	19.6	19.0	23.0	16.5	18.0	5.0	10.3	1.2	3.0	1.3	13.1	7.9	9.1	5.6	7.0	16.6	13.5	4.8	4.9	10.2	11.2	

(Continued on next page)

TABLE III  
(Continued)

Site	Year	mm		[NH <sub>4</sub> <sup>+</sup> ]		[NO <sub>3</sub> <sup>-</sup> ]		[SO <sub>4</sub> <sup>2-</sup> ]		[Ca <sup>2+</sup> ]		[Mg <sup>2+</sup> ]		[K <sup>+</sup> ]		[Na <sup>+</sup> ]		[Cl <sup>-</sup> ]		[H <sup>+</sup> ]		Median pH		Conductivity		
		BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP
BEA	00	1195	1025	24.4	27.1	24.0	42.2	18.6	20.5	29.2	40.3	3.7	8.5	2.8	18.9	8.0	11.9	7.5	13.7	3.3	7.8	5.8	5.2	9.2	15.2	
	01	1545	1283	19.0	18.9	16.6	29.9	14.2	15.3	18.7	32.6	2.7	7.1	2.6	15.7	5.1	8.1	4.0	7.5	3.2	4.6	5.9	5.5	6.9	11.4	
SCH	99	2440	2119	32.9	36.8	35.0	44.8	22.9	28.1	18.8	46.5	3.2	15.0	2.9	38.2	11.0	13.4	7.0	12.7	9.7	6.4	5.3	5.5	10.6	16.9	
	00	2387	2002	33.7	43.2	29.9	44.0	22.3	28.3	22.2	54.6	3.8	14.9	4.6	30.7	7.9	11.7	8.0	13.0	3.0	2.0	5.8	6.2	10.6	19.1	
Alps	01	2014	1700	31.5	37.1	26.4	39.4	18.9	25.8	18.8	57.6	3.5	18.5	3.9	37.3	7.3	9.2	5.9	12.4	4.9	2.1	5.8	6.4	9.3	17.4	
	00	1153	1114	12.2	9.4	17.5	18.0	17.9	20.4	40.6	39.1	4.4	8.9	3.2	12.8	11.3	13.0	10.3	10.6	6.7	6.4	5.8	5.6	9.4	11.2	
CEL	01	761	684	20.2	12.8	18.6	16.1	18.2	18.9	22.7	30.0	3.8	10.0	2.9	10.7	5.6	7.8	5.9	6.4	3.6	4.0	5.6	5.5	8.2	9.8	
	99	1037	939	18.9	15.9	26.2	22.3	18.0	15.8	30.9	35.0	6.1	10.5	1.6	5.9	10.6	11.4	8.7	7.5	4.3	4.6	6.0	5.6	7.4	8.4	
NAT	00	1245	1170	12.8	9.9	16.4	18.2	16.9	16.4	32.9	35.3	5.4	10.4	2.5	6.4	5.3	6.0	8.9	7.4	5.1	4.5	5.6	5.5	8.5	9.1	
	01	893	812	15.2	13.4	17.2	18.1	14.5	15.3	21.7	29.6	5.7	11.5	2.7	7.3	5.2	6.1	5.5	4.6	2.9	3.7	6.0	5.7	7.0	8.5	
Southern Alps																										
NOV	97	1654	1401	44.8	44.1	43.6	62.0	42.7	50.6	14.4	32.4	4.1	17.3	2.5	31.9	11.4	14.4	10.5	14.8	28.9	18.5	4.6	4.8	19.2	24.1	
	98	2126	1885	52.1	55.3	47.6	67.5	47.0	53.4	16.8	34.7	3.7	13.5	1.8	20.9	11.4	12.4	10.5	12.2	29.9	23.5	4.5	4.5	20.9	24.4	
CHI**B23	99	2181	1906	58.5	60.3	48.9	70.2	48.1	56.7	31.9	50.6	6.3	19.6	3.2	40.4	19.2	23.4	14.9	17.9	12.1	10.2	5.3	5.5	17.8	24.3	
	00	2771	2435	40.0	38.5	38.5	54.2	36.4	41.2	31.6	46.7	4.7	15.6	3.3	27.8	11.6	15.0	10.7	13.6	10.9	9.6	5.2	5.2	16.8	20.6	
CHI**B23	01	1728	1584	57.7	50.8	49.4	67.5	47.1	53.2	33.2	55.1	6.3	19.4	2.8	41.5	11.0	15.3	9.9	14.4	9.2	7.1	5.1	5.9	18.9	24.3	
	01	1865	1768	33.6	30.5	30.5	31.5	29.5	29.5	29.5	29.5	4.9	4.9	2.7	6.9	6.9	5.0	5.0	5.9	5.9	5.7	5.7	5.7	5.7	12.8	

\*Means of both subplots.

\*\*Precipitation amount, nitrogen and sulfate concentrations in winter under the forest canopy were estimated (see text).

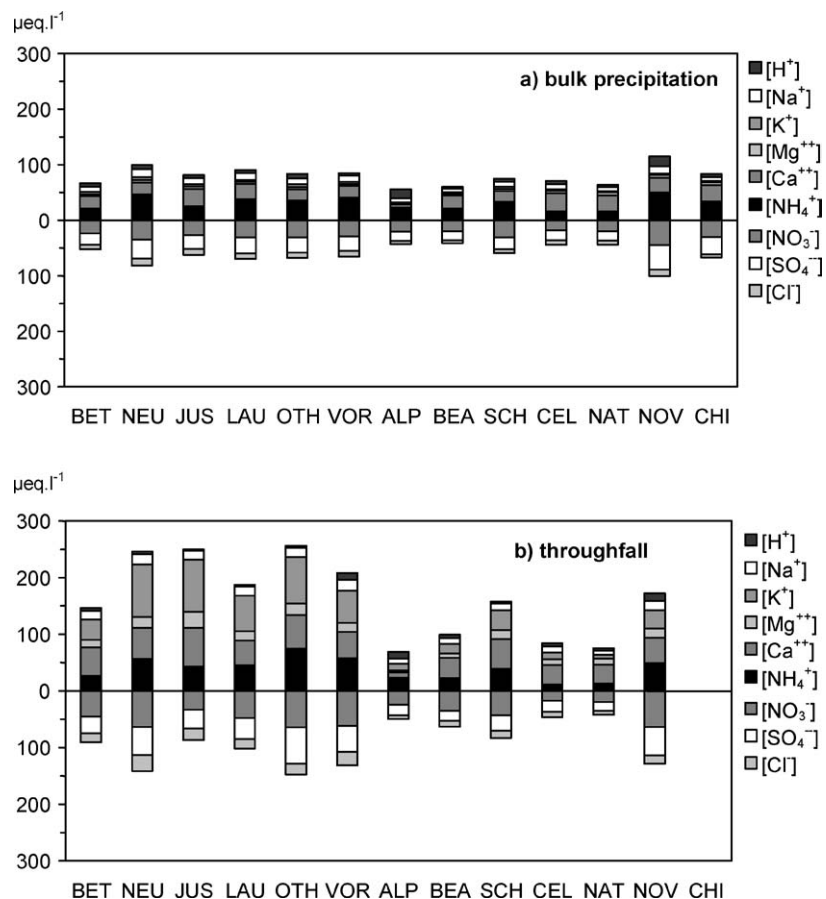


Figure 2. Volume-weighted mean concentrations in bulk precipitation (a) and throughfall (b) (in  $\mu\text{eq l}^{-1}$ ) on LWF plots. Cations are plotted on the upper part of the y-axis, and anions on the lower part. Variations in concentrations related, e.g., to the time of the year or to the precipitation, amounts during the sampling interval may be significant but are not dealt with in this paper.

### 428 3.3. FLUXES

429 Nitrogen and sulfur fluxes in bulk precipitation and throughfall (Table IV, Figure 3)  
 430 were highest at NOV in the Southern Alps (on average  $\text{BP}_\text{N} = 27.9 \text{ kg ha}^{-1} \text{ a}^{-1}$ ,  
 431  $\text{TF}_\text{N} = 29.2 \text{ kg ha}^{-1} \text{ a}^{-1}$ ,  $\text{BP}_\text{S} = 14.7 \text{ kg ha}^{-1} \text{ a}^{-1}$ ,  $\text{TF}_\text{S} = 14.9 \text{ kg ha}^{-1} \text{ a}^{-1}$ ),  
 432 followed by SCH in the Lower Alps. They were lowest at the high-elevation sites  
 433 (CEL, NAT), where  $\text{BP}_\text{N} = 4.9 \text{ kg ha}^{-1} \text{ a}^{-1}$ ,  $\text{TF}_\text{N} = 4.0 \text{ kg ha}^{-1} \text{ a}^{-1}$ ,  $\text{BP}_\text{S} =$   
 434  $2.8 \text{ kg ha}^{-1} \text{ a}^{-1}$ ,  $\text{TF}_\text{S} = 2.6 \text{ kg ha}^{-1} \text{ a}^{-1}$  on average at both plots. The throughfall  
 435 deposition of N was higher than its bulk deposition on all plots except the two  
 436 plots at high elevations (CEL, NAT). The difference between throughfall and bulk  
 deposition fluxes was highest at OTH (on average  $5.7 \text{ kg N ha}^{-1} \text{ a}^{-1}$ ), which was

TABLE IV  
Annual element fluxes ( $\text{kg ha}^{-1} \text{a}^{-1}$ ) in the open field (bulk precipitation BP) and under the canopy (throughfall TF)

Site	Year	$\text{NH}_4\text{-N}$		$\text{NO}_3\text{-N}$		Inorg.-N		$\text{SO}_4\text{-S}$		Ca		Mg		K		Na		Cl		H	
		BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF	BP	TF
Jura	BET	5.4	6.2	6.7	10.8	12.2	17.0	5.9	7.5	7.1	15.2	0.7	2.5	2.8	21.7	5.1	6.1	5.0	9.2	0.18	0.14
	00	4.0	4.3	4.3	7.1	8.2	11.4	4.6	6.0	6.1	12.2	0.5	2.0	2.0	17.9	2.7	4.5	4.2	7.4	0.06	0.05
	01	4.2	4.7	4.2	7.7	8.4	12.4	4.6	6.0	6.8	13.4	0.6	2.0	2.0	18.0	2.4	3.3	3.6	5.7	0.05	0.03
	NEU	5.1	5.1	3.8	5.8	9.0	10.8	4.3	5.2	3.4	7.2	0.5	1.5	1.5	23.4	2.7	2.8	3.7	6.3	0.06	0.03
Central Plateau	JUS	3.0	4.2	3.3	2.8	6.3	7.0	3.7	4.0	4.0	8.6	0.4	2.8	1.8	31.1	2.2	2.6	2.9	5.8	0.10	0.02
	99	5.2	7.7	6.0	6.1	11.2	13.8	5.5	6.1	8.5	15.7	0.8	3.7	1.6	39.8	4.6	5.0	6.6	8.5	0.07	0.03
	00	3.6	3.9	4.0	3.3	7.7	7.2	3.6	4.0	7.3	11.3	0.7	3.0	1.6	25.6	2.6	2.8	4.9	5.7	0.03	0.01
	01	3.7	5.2	3.8	3.7	7.5	8.9	4.4	4.6	8.1	11.3	0.7	2.6	1.7	28.0	2.2	2.2	3.1	4.8	0.03	0.01
	97	7.5	7.5	5.7	7.8	13.2	15.3	6.8	7.7	5.9	8.6	0.6	2.1	2.3	28.9	3.2	3.4	4.6	6.3	0.12	0.05
	98	6.4	7.6	5.4	7.7	11.9	15.3	6.0	6.9	4.7	9.1	0.6	2.5	1.4	24.9	3.7	3.9	4.7	7.1	0.11	0.07
	99	8.5	7.8	7.1	9.8	15.6	17.7	7.1	7.9	11.3	12.8	0.8	2.9	1.6	41.0	5.7	6.6	6.4	8.6	0.06	0.07
	00	6.8	7.2	6.0	7.2	12.8	14.4	5.6	6.0	7.4	9.4	0.7	1.9	1.6	21.3	3.1	3.6	5.1	6.6	0.05	0.02
	01	6.8	6.4	5.2	6.1	12.1	12.5	5.5	5.9	7.5	10.3	0.7	2.0	1.8	25.3	2.6	3.0	3.2	5.2	0.03	0.02
	95	5.6	8.4	4.9	7.2	10.6	15.6	5.7	10.2	3.4	10.2	0.4	2.7	1.6	30.9	1.9	2.6	2.4	5.2	0.15	0.03
	96	6.2	9.1	5.3	8.1	11.5	17.2	5.6	10.1	3.1	9.5	0.4	2.2	1.2	27.5	2.5	3.0	3.2	4.9	0.12	0.04
	97	4.4	7.6	3.6	6.4	8.1	14.0	3.9	7.4	2.6	7.6	0.4	1.6	2.2	22.4	1.9	2.5	3.0	4.7	0.09	0.02
98	4.9	9.7	4.0	7.2	8.9	16.9	4.7	8.4	4.0	8.6	0.6	1.9	3.7	25.4	3.1	3.5	4.9	6.7	0.12	0.03	
99	6.3	9.5	6.3	8.5	12.5	18.0	5.6	9.2	6.8	11.8	0.9	2.6	3.4	36.5	4.3	5.0	5.4	6.5	0.07	0.02	
00	5.8	8.4	5.0	7.9	10.8	16.3	4.9	7.3	6.4	9.5	0.5	1.5	1.4	17.2	2.7	3.1	4.0	5.7	0.04	0.03	
01	5.2	7.4	4.1	6.0	9.3	13.4	4.5	6.7	5.5	10.1	0.6	1.8	2.2	23.4	2.1	2.6	2.9	4.7	0.03	0.01	
97	6.3	6.6	4.3	6.4	10.6	12.9	4.3	6.4	2.8	6.2	0.4	1.4	2.0	21.7	2.4	3.1	3.0	7.0	0.07	0.20	
98	6.8	6.5	4.5	7.2	11.3	13.7	5.1	7.1	4.2	7.3	0.5	1.7	1.4	19.6	3.4	4.1	4.6	8.0	0.08	0.11	
99	8.4	7.6	7.0	10.0	15.3	17.6	5.7	7.6	7.7	10.9	0.9	2.0	1.8	17.9	4.9	5.4	6.7	9.1	0.08	0.12	
00	6.3	8.4	4.6	8.3	11.0	16.7	4.5	6.0	4.8	8.3	0.4	1.4	1.3	13.0	2.6	3.4	4.5	6.0	0.03	0.04	
01	6.7	6.0	4.7	5.9	11.3	11.9	4.7	5.3	5.8	8.7	0.6	1.6	1.9	25.1	2.4	3.1	3.5	6.9	0.02	0.09	



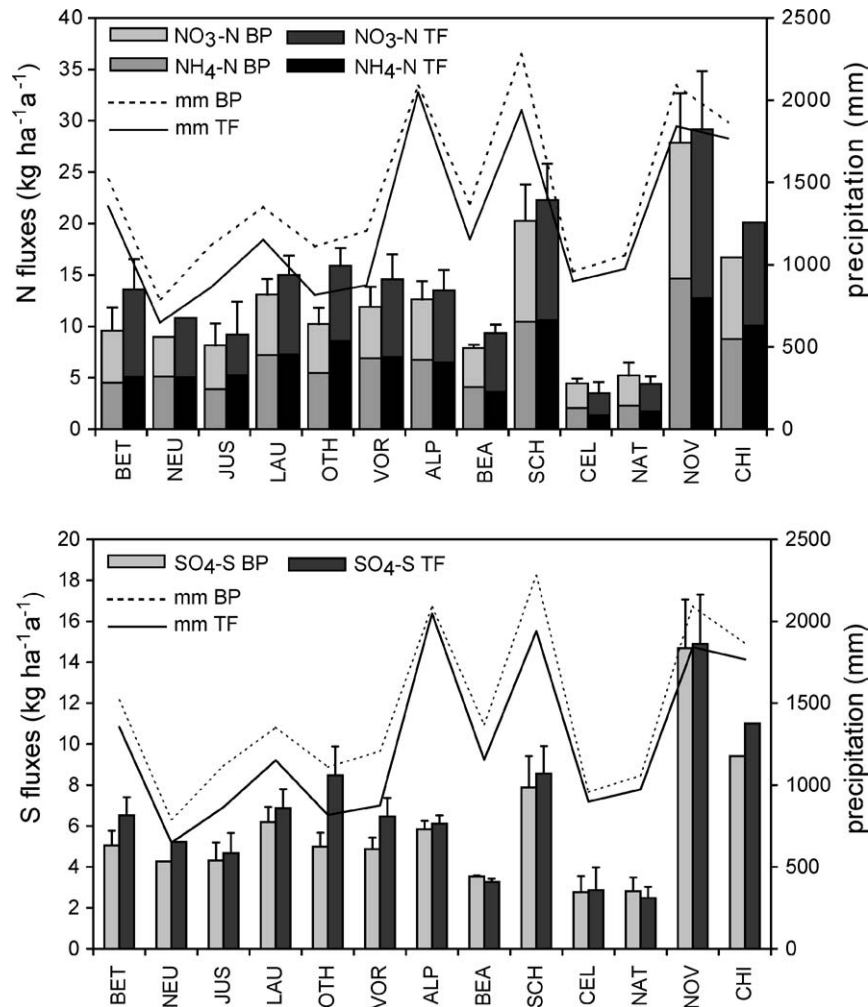


Figure 3. Mean annual inorganic nitrogen ( $\text{NH}_4\text{-N} + \text{-N}$ ) (top) and sulfate ( $\text{SO}_4\text{-S}$ ) (bottom) fluxes ( $\text{kg ha}^{-1} \text{a}^{-1}$ ) in the open area (BP) and in the forest stand (TF). Mean annual precipitation in the open area and in the stand ( $\text{mm a}^{-1}$ ) is also shown.

also the plot with the highest interception. It was rather low for the other plots. The 437  
deposition pattern for S was similar to the pattern for N. Two plots (NAT, BEA) 438  
showed lower  $\text{SO}_4^{2-}$  fluxes in throughfall than in bulk deposition. 439

Bulk deposition of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  was highest at NOV, and lowest at ALP 440  
(Table IV). Throughfall enrichment in  $\text{Ca}^{2+}$  was highest at SCH and BET, and 441  
throughfall deposition of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  was highest in NOV and SCH. It was 442  
lowest at ALP. The year-to-year variation in  $\text{Ca}^{2+}$  deposition could be quite large 443  
(NOV, CEL).  $\text{Na}^+$  and  $\text{Cl}^-$  fluxes in both bulk precipitation and throughfall were 444  
highest in NOV. 445

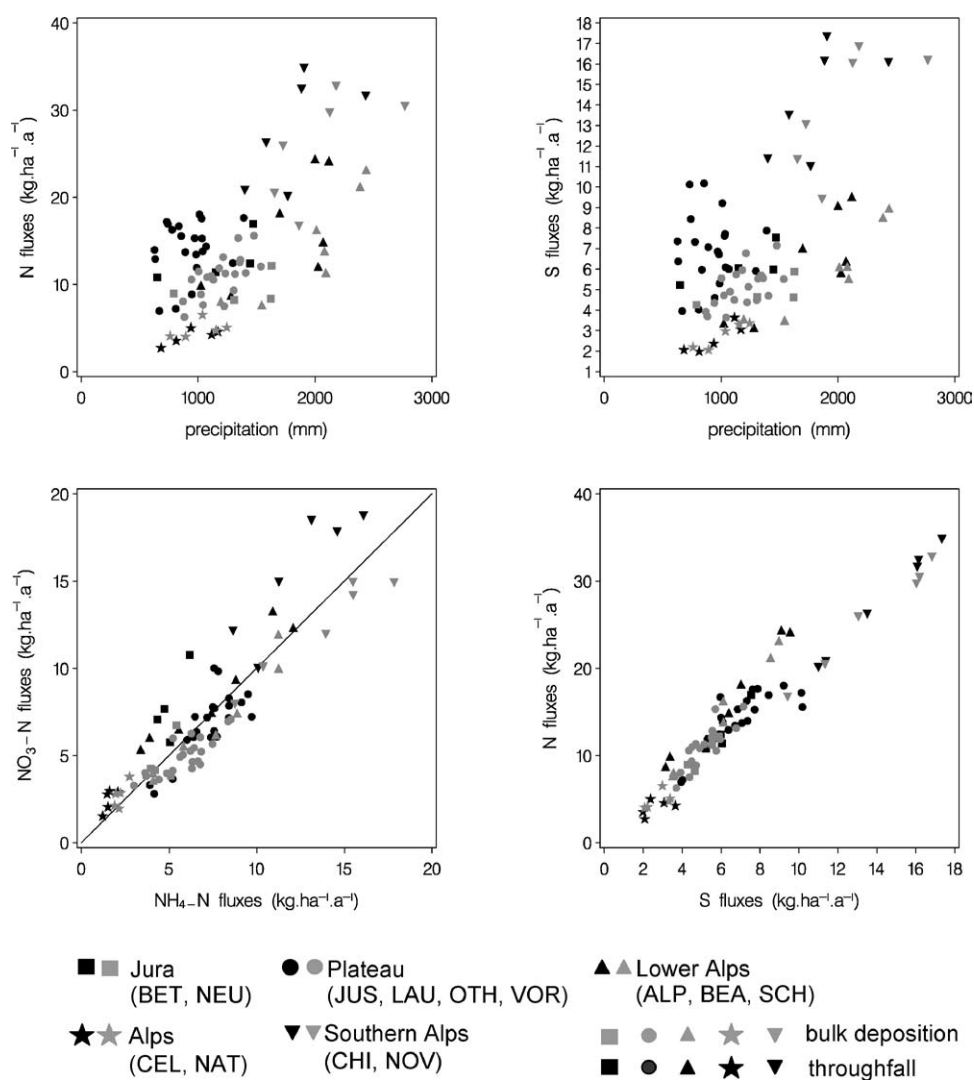


Figure 4. Relationships on each LWF plot between: (a) annual inorganic nitrogen ( $\text{NH}_4\text{-N} + \text{NO}_3\text{-N}$ ) fluxes and annual precipitation in the open area and in the forest stand; (b) annual sulfate ( $\text{SO}_4\text{-S}$ ) fluxes and annual precipitation in the open area and in the forest stand; (c)  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  annual fluxes; (d) N and S annual fluxes.

446 The annual N and  $\text{SO}_4^{2-}$  depositions positively correlated with the annual pre-  
 447 cipitation amounts (Figure 4) at the regional level (plot-to-plot variation), as well  
 448 as at the plot level (year-to-year variation for each plot). Plots with high annual  
 449 precipitation tended to have higher bulk deposition fluxes, even though the differ-  
 450 ences in pollution climate between plots were still apparent for plots with similar  
 451 precipitation amounts but different element fluxes (e.g. NOV, SCH and ALP). The  
 452 correlation was stronger for bulk deposition than for throughfall. N and  $\text{SO}_4^{2-}$

concentrations were closely correlated, as were N and  $\text{SO}_4^{2-}$  depositions.  $\text{NH}_4^+$  and  $\text{NO}_3^-$  concentrations and depositions were also significantly correlated. The contribution of  $\text{NH}_4^+$  to inorganic N deposition varied from 41% (CEL in 2000) to 61% (VOR in 1998) in bulk deposition and from 34% (CEL in 2000) to 60% (JUS in 1998) in throughfall.

### 3.4. ESTIMATES OF THE COMPONENTS OF ATMOSPHERIC DEPOSITION COMPARED WITH MODELED DEPOSITION

The estimated wet and dry components of atmospheric deposition, as well as the estimated canopy leaching or uptake, are given in Table V for each sampling year. Negative dry  $\text{SO}_4^{2-}$  deposition estimates may have to be interpreted as  $\text{SO}_4^{2-}$  uptake from the canopy (NAT in 1999). NOV was clearly the plot with the highest total loads of N and S, followed by SCH.

Estimated wet deposition of N (corrected bulk deposition,  $\text{WD}_{\text{N(LWF)}}$ ) and modeled wet deposition ( $\text{WD}_{\text{N(mod)}}$ ) were closely correlated ( $r = 0.87$ ,  $p < 0.001$ ,  $n = 13$ ; Table VI, Figure 5). The highest difference between modeled and LWF values were observed at ALP, where  $\text{WD}_{\text{N(mod)}}$  exceeded  $\text{WD}_{\text{N(LWF)}}$  by  $6.3 \text{ kg N ha}^{-1} \text{ a}^{-1}$ . The LWF estimates for dry deposition of N ( $\text{DD}_{\text{N(LWF)}}$ ) also correlated with the modeled values ( $\text{DD}_{\text{N(mod)}}$ ), though less closely ( $r = 0.70$ ,  $p < 0.05$ ,  $n = 12$ ). The correlation between LWF total deposition ( $\text{TD}_{\text{N(LWF)}}$ ) and modeled total deposition ( $\text{TD}_{\text{N(mod)}}$ ) was highly significant ( $r = 0.84$ ,  $p < 0.001$ ,  $n = 12$ ).  $\text{DD}_{\text{N(mod)}}$ , however, systematically exceeded  $\text{DD}_{\text{N(LWF)}}$  and  $\text{TD}_{\text{N(mod)}}$   $\text{TD}_{\text{N(LWF)}}$ , especially at ALP, with the exception of NOV.

The correlation between sulfur WD derived from BP values ( $\text{WD}_{\text{S(LWF)}}$ ) and the modeled deposition for S ( $\text{WD}_{\text{S(mod)}}$ ) was also significant ( $r = 0.75$ ,  $p < 0.01$ ,  $n = 13$ ; Table VI, Figure 5). The main differences between  $\text{WD}_{\text{S(LWF)}}$  and  $\text{WD}_{\text{S(mod)}}$  were observed at ALP ( $\text{WD}_{\text{S(mod)}}$  exceeded  $\text{WD}_{\text{S(LWF)}}$  by  $2.9 \text{ kg ha}^{-1} \text{ a}^{-1}$ ) and NOV ( $\text{WD}_{\text{S(mod)}}$  was lower than  $\text{WD}_{\text{S(LWF)}}$  by  $5.2 \text{ kg ha}^{-1} \text{ a}^{-1}$ ). The correlation between LWF dry deposition for S ( $\text{DD}_{\text{S(LWF)}}$ ) and modeled dry deposition ( $\text{DD}_{\text{S(mod)}}$ ) was somewhat weaker ( $r = 0.69$ ,  $p < 0.05$ ,  $n = 12$ ), as was the correlation between LWF and modeled total deposition for S ( $r = 0.70$ ,  $p < 0.05$ ,  $n = 12$  between  $\text{TD}_{\text{S(LWF)}}$  and  $\text{TD}_{\text{S(mod)}}$ ).  $\text{DD}_{\text{S(mod)}}$  exceeded  $\text{DD}_{\text{S(LWF)}}$  for the four plots located on the Plateau and NEU.  $\text{TD}_{\text{S(mod)}}$  was higher than  $\text{TD}_{\text{S(LWF)}}$  for all plots except NOV.

### 3.5. CRITICAL LOADS FOR NITROGEN

The SMB approach yielded critical loads for N, which was within the range of critical loads empirically established for the managed plots (Table VII). On unmanaged plots, where no N is removed by wood harvesting ( $U_{\text{N}} = 0$ ), the SMB critical loads were lower than the empirical ones.  $\text{TD}_{\text{N(mod)}}$  exceeded the SMB critical loads on

TABLE V  
 Estimated components of atmospheric deposition (WD, DD), canopy uptake (CU) of nitrogen and canopy leaching (Cle) of base cations  
 (in kg ha<sup>-1</sup>a<sup>-1</sup>)

Site	Year	NH <sub>4</sub> -N			NO <sub>3</sub> -N			Inorg.-N			S			Ca			Mg			K			Na			Cl			
		WD	DD	CU	WD	DD	CU	WD	DD	CU	WD	DD	TD	WD	DD	Cle	WD	DD	Cle	WD	DD	Cle	WD	DD	DD	WD	DD	DD	
Jura																													
BET	1999	4.8	4.5	3.1	5.6	6.1	0.9	10.4	10.6	4.1	21.0	4.9	2.6	4.9	3.7	6.6	0.5	0.3	1.6	1.9	1.5	18.3	4.1	2.0	4.1	5.2			
	2000	3.5	2.0	1.2	3.5	3.8	0.3	7.1	5.8	1.5	12.9	3.9	2.1	4.2	5.8	2.1	0.4	0.5	1.1	1.4	2.0	14.6	2.2	2.3	3.4	4.0			
	2001	3.7	4.7	3.8	3.5	5.2	1.0	7.2	10.0	4.8	17.2	3.9	2.1	4.7	4.5	4.3	0.5	0.4	1.2	1.3	1.4	15.3	2.0	1.3	2.9	2.8			
NEU	1998	4.5	4.5	4.0	3.2	3.3	0.8	7.7	7.8	4.7	15.6	3.6	1.6	2.3	1.1	3.7	0.4	0.1	1.0	1.0	0.5	21.9	2.2	0.6	3.0	3.3			
	2001	4.5	4.5	4.0	3.2	3.3	0.8	7.7	7.8	4.7	15.6	3.6	1.6	2.3	1.1	3.7	0.4	0.1	1.0	1.0	0.5	21.9	2.2	0.6	3.0	3.3			
Central Plateau																													
JUS	1998	2.7	2.8	1.4	2.7	0.2	0.2	5.4	3.1	1.5	8.5	3.1	0.8	2.8	2.0	3.8	0.3	0.2	2.3	1.2	1.0	28.9	1.8	0.8	2.4	3.4			
	1999	4.7	4.3	1.2	5.0	1.3	0.2	9.6	5.6	1.4	15.2	4.7	1.4	5.9	3.3	6.5	0.5	0.3	2.9	1.1	0.7	38.1	3.7	1.2	5.4	3.1			
2000	2000	3.2	2.3	1.6	3.3	0.2	0.2	6.6	2.5	1.8	9.1	3.1	1.0	5.0	2.9	3.4	0.5	0.2	2.3	1.1	0.7	23.8	2.1	0.7	4.0	1.7			
	2001	3.3	4.7	2.8	3.2	0.8	0.3	6.5	5.6	3.2	12.0	3.7	0.9	5.6	2.5	3.2	0.5	0.2	1.9	1.2	0.6	26.3	1.8	0.4	2.6	2.2			
LAU	1997	6.7	5.5	4.7	4.7	3.9	0.8	11.4	9.4	5.5	20.8	5.7	2.0	4.1	2.3	2.2	0.4	0.2	1.5	1.5	0.9	26.4	2.6	0.8	3.8	2.5			
	1998	5.7	4.9	3.1	4.5	3.7	0.5	10.2	8.6	3.6	18.9	5.0	1.9	3.3	1.8	4.0	0.5	0.2	1.8	1.0	0.6	23.4	3.0	0.9	3.9	3.2			
1999	1999	7.6	5.8	5.5	5.9	5.1	1.2	13.5	10.9	6.7	24.4	6.0	1.9	7.8	5.2	(-0.2)	0.6	0.4	1.9	1.1	0.8	39.2	4.6	2.0	5.2	3.4			
	2000	6.0	4.3	3.2	5.0	2.7	0.5	11.1	7.0	3.7	18.1	4.7	1.3	5.1	3.4	0.9	0.5	0.3	1.2	1.0	0.7	19.5	2.5	1.1	4.2	2.5			
2001	2001	6.1	4.6	4.3	4.3	2.4	0.7	10.4	7.0	5.0	17.5	4.6	1.3	5.2	3.6	1.5	0.5	0.3	1.3	1.2	0.9	23.2	2.1	0.9	2.6	2.6			
	1995	5.0	7.9	4.5	4.1	3.7	0.6	9.1	11.6	5.1	20.7	4.8	5.4	2.3	2.2	5.6	0.3	0.2	2.2	1.0	1.1	28.8	1.6	1.0	2.0	3.2			
OTH	1996	5.5	8.1	4.5	4.4	4.3	0.7	9.9	12.4	5.1	22.3	4.7	5.5	2.1	1.5	5.8	0.3	0.2	1.7	0.8	0.6	26.0	2.1	0.9	2.6	2.3			
	1997	3.9	7.8	4.2	3.0	4.0	0.6	7.0	11.8	4.7	18.7	3.3	4.1	1.8	1.5	4.3	0.3	0.2	1.1	1.5	1.3	19.6	1.6	0.9	2.5	2.3			
1998	1998	4.3	10.1	4.7	3.3	4.5	0.6	7.7	14.6	5.3	22.2	4.0	4.5	2.8	1.8	4.0	0.4	0.2	1.3	2.5	1.7	21.2	2.5	1.0	4.0	2.7			
	1999	5.6	8.5	4.5	5.2	4.0	0.7	10.8	12.5	5.2	23.3	4.7	4.5	4.7	3.1	4.0	0.7	0.4	1.6	2.3	1.6	32.6	3.5	1.5	4.4	2.1			
2000	2000	5.2	6.7	3.4	4.2	4.2	0.5	9.3	10.9	4.0	20.2	4.1	3.2	4.4	3.1	2.0	0.4	0.2	0.9	0.9	0.7	15.7	2.2	1.0	3.3	2.3			
	2001	4.6	6.9	4.1	3.4	3.2	0.6	8.1	10.1	4.7	18.1	3.8	2.9	3.8	2.8	3.5	0.4	0.3	1.1	1.5	1.1	20.8	1.7	0.8	2.4	2.3			
VOR	1997	5.6	2.3	1.4	3.5	3.0	0.2	9.2	5.4	1.6	14.5	3.7	2.7	1.9	1.6	2.7	0.3	0.2	0.9	1.4	1.2	19.2	2.0	1.1	2.4	4.6			
	1998	6.0	3.5	3.0	3.7	4.1	0.6	9.8	7.5	3.6	17.3	4.3	2.8	2.9	2.2	2.2	0.4	0.2	1.1	0.9	0.8	18.0	2.7	1.3	3.8	4.2			
1999	1999	7.5	4.4	4.3	5.8	5.2	0.9	13.2	9.5	5.2	22.8	4.8	2.8	5.3	3.2	2.4	0.6	0.3	1.0	1.2	0.8	15.9	4.0	1.4	5.5	3.6			
	2000	5.6	5.6	2.9	3.8	4.9	0.5	9.5	10.5	3.3	20.0	3.8	2.2	3.3	3.1	1.9	0.3	0.3	0.8	0.9	0.8	11.3	2.1	1.3	3.7	2.3			
2001	5.9	2.0	1.9	3.9	2.3	0.3	9.8	4.3	2.2	14.1	4.0	1.4	4.0	3.5	1.2	0.4	0.3	0.9	1.3	1.2	22.6	1.9	1.1	2.8	4.0				

(Continued on next page)

TABLE V  
(Continued)

Site	Year	NH <sub>4</sub> - N		NO <sub>3</sub> - N		Inorg. N		S		Ca		Mg		K		Na		Cl								
		WD	DD	CU	DD	CU	TD	WD	DD	WD	DD	WD	DD	WD	DD	WD	DD	WD	DD	WD	DD					
Lower Alps																										
ALP	1997	6.8	2.7	2.1	5.2	2.7	0.4	12.0	5.4	2.5	17.4	5.2	1.2	1.8	0.9	1.7	0.2	0.1	0.3	1.0	0.5	7.4	3.2	0.9	3.3	1.1
	1998	5.2	1.5	1.1	4.6	2.1	0.2	9.8	3.6	1.3	13.4	4.7	1.2	1.4	0.9	1.9	0.2	0.1	0.4	0.7	0.5	9.2	3.1	1.2	3.4	1.6
BEA	2000	3.6	1.6	1.4	3.3	3.1	0.4	7.0	4.7	1.7	11.7	3.0	0.4	4.8	4.1	(-0.6)	0.4	0.3	0.4	0.9	0.8	5.9	1.8	1.0	2.6	2.4
	2001	3.7	1.4	1.7	3.0	2.8	0.4	6.6	4.2	2.1	10.9	3.0	0.2	4.0	3.7	0.7	0.4	0.3	0.4	1.1	1.0	5.8	1.5	0.9	1.8	1.6
SCH	1999	10.0	5.2	4.3	9.9	4.2	0.9	19.9	9.5	5.2	29.4	7.5	2.0	6.3	3.4	10.0	0.7	0.3	2.9	1.9	1.1	28.7	5.0	1.5	5.0	4.5
	2000	10.0	6.2	4.1	8.3	4.7	0.7	18.3	10.9	4.8	29.3	7.2	1.9	7.3	5.9	8.6	0.8	0.6	2.3	2.9	2.5	18.7	3.5	1.9	5.5	3.7
	2001	7.9	8.0	7.1	6.2	4.5	1.3	14.1	12.5	8.4	26.6	5.1	1.9	5.2	2.8	11.5	0.6	0.3	2.9	2.1	1.2	21.5	2.7	0.9	3.5	4.0
Alps																										
CEL	2000	1.8	0.7	1.0	2.3	0.8	0.3	4.1	1.5	1.3	5.6	2.8	0.9	6.5	4.0	(-1.7)	0.5	0.2	0.5	1.0	0.6	3.9	2.4	0.9	3.5	0.7
	2001	1.9	(-0.3)	0.4	1.6	(-0.0)	0.1	3.6	(-0.3)	0.5	3.2	1.9	0.2	2.4	2.0	(-0.2)	0.3	0.2	0.4	0.6	0.5	1.8	0.8	0.4	1.3	0.3
NAT	1999	2.4	0.4	0.7	3.2	(-0.1)	0.2	5.6	0.3	0.9	5.9	2.5	(-0.1)	4.4	1.8	0.4	0.6	0.2	0.5	0.4	0.2	1.5	2.1	0.4	2.6	(-0.1)
	2000	2.0	0.6	1.0	2.4	0.9	0.3	4.4	1.5	1.3	5.9	2.8	0.2	5.7	3.0	(-0.4)	0.6	0.3	0.6	0.8	0.5	1.6	1.2	0.4	3.2	(-0.2)
	2001	1.7	0.5	0.7	1.8	0.4	0.1	3.5	0.9	0.8	4.4	1.7	0.2	2.7	1.4	0.7	0.5	0.2	0.5	0.6	0.4	1.3	0.9	0.3	1.4	(-0.1)
Southern Alps																										
NOV	1997	9.2	2.0	2.5	8.4	4.4	0.6	17.6	6.3	3.1	24.0	9.5	1.9	3.3	1.8	4.0	0.6	0.3	2.1	1.1	0.6	15.8	3.5	1.1	5.0	2.3
	1998	13.8	3.9	3.1	11.8	6.7	0.6	25.6	10.6	3.7	36.1	13.5	2.7	4.9	2.0	6.2	0.7	0.2	2.2	1.0	0.4	13.9	4.5	0.9	6.5	1.7
	1999	15.9	6.0	5.8	12.4	7.5	1.1	28.3	13.5	7.0	41.8	14.1	3.2	9.6	5.3	4.4	1.2	0.6	2.7	1.9	1.1	27.2	7.8	2.5	9.5	2.6
	2000	13.8	3.7	4.4	12.4	7.1	1.0	26.2	10.8	5.4	37.0	13.6	2.5	12.1	7.9	2.8	1.2	0.6	2.8	2.4	1.7	22.4	6.0	2.4	8.6	3.1
	2001	12.4	2.8	4.0	9.9	5.9	0.9	22.4	8.8	4.9	31.1	11.0	2.5	7.9	6.7	2.8	1.0	0.7	2.1	1.3	1.2	23.2	3.6	2.0	5.0	3.2
CHI	2001	7.8	—	—	6.6	—	—	14.4	—	—	—	7.9	3.1	7.6	—	—	0.8	—	—	1.3	—	—	2.4	—	2.7	—

TABLE VI

Mean annual nitrogen ( $\text{NO}_3\text{-N} + \text{NH}_4\text{-N}$ ) and sulfur fluxes based on measurements on the LWF plots (wet deposition WD, throughfall TF, estimated dry deposition DD and total deposition TD) and modeled wet deposition (WD) of nitrogen ( $\text{NO}_3\text{-N} + \text{NH}_4\text{-N}$ ) and sulfate, modeled DD of nitrogen ( $\text{NO}_x$  and  $\text{NH}_y$ ) and sulfur dioxide ( $\text{SO}_2$ )

	N ( $\text{NO}_3\text{-N} + \text{NH}_4\text{-N}$ ) fluxes ( $\text{kg N ha}^{-1}\text{a}^{-1}$ )						S fluxes ( $\text{kg S ha}^{-1}\text{a}^{-1}$ )							
	LWF			Modeled deposition			LWF			Modeled deposition				
	WD	DD	TD	TF	TD/TF	WD	DD	TD	WD	DD	TD	WD	DD	TD
Jura														
BET	8.2	8.8	17.0	13.6	1.3	11.7	9.9	21.6	4.2	2.3	6.5	5.3	2.2	7.5
NEU	7.7	7.8	15.6	10.8	1.4	8.1	9.6	17.8	3.6	1.6	5.2	3.7	5.0	8.7
Central Plateau														
JUS	7.0	4.2	11.2	9.2	1.2	7.6	10.1	17.7	3.6	1.0	4.7	3.4	4.7	8.2
LAU	11.3	8.6	19.9	15.0	1.3	9.4	14.4	23.8	5.2	1.7	6.9	4.3	2.7	7.0
OTH	8.8	12.0	20.8	15.9	1.3	8.8	19.6	28.4	4.2	4.3	8.5	4.0	5.8	9.8
VOR	10.2	7.5	17.8	14.6	1.2	8.7	19.9	28.6	4.1	2.4	6.5	3.9	5.3	9.2
Lower Alps														
ALP	10.9	4.5	15.4	13.5	1.1	17.2	11.1	28.3	4.9	1.2	6.1	7.8	1.3	9.2
BEA	6.8	4.5	11.3	9.4	1.2	8.9	9.6	18.5	3.0	0.3	3.3	4.1	0.4	4.5
SCH	17.3	11.1	28.4	22.3	1.3	14.0	13.9	27.9	6.6	2.0	8.6	6.4	3.2	9.5
Alps														
CEL	3.7	0.7	4.4	3.5	1.3	6.4	5.6	12.0	2.3	0.5	2.8	3.0	0.7	3.7
NAT	4.5	0.9	5.4	4.4	1.2	6.4	3.2	9.6	2.4	0.1	2.5	3.0	0.8	3.8
S. Alps														
NOV	24.0	10.0	34.0	29.2	1.2	20.8	8.8	29.6	12.3	2.6	14.9	7.1	2.2	9.4
CHI	13.9			20.1		13.5	7.5	21.0	7.6	3.4	11.0	7.2	2.1	9.3

*Note.* Years of reference for modeled nitrogen deposition: 2000 north of the Alps, 1993–2000 south of the Alps.

Years of reference for modeled wet deposition of sulfur: 1997–1999; for dry deposition of  $\text{SO}_2$ : 2000. Estimated LWF WD: BP\* correction factor.

Estimated LWF TD of N: TD after the canopy budget model.

Estimated LWF Deposition of S: TD = TF, DD = TD-WD.

491 all plots, whereas  $\text{TD}_{\text{N(LWF)}}$  was below the SMB critical load on the two alpine  
 492 plots (CEL, NAT), ALP, and the managed subplot of JUS. NOV, SCH, and, to a  
 493 lesser extent, CHI were the three plots where throughfall, which provides a lower  
 494 estimate of total N deposition, exceeded both SMB and empirical critical loads.

495

#### 4. Discussion

496 4.1. REGIONAL DISTRIBUTION OF ATMOSPHERIC DEPOSITION

497 At the high-elevation sites (CEL and NAT), which are remote from pollution  
 498 sources, measured atmospheric deposition was low (only a little higher than what

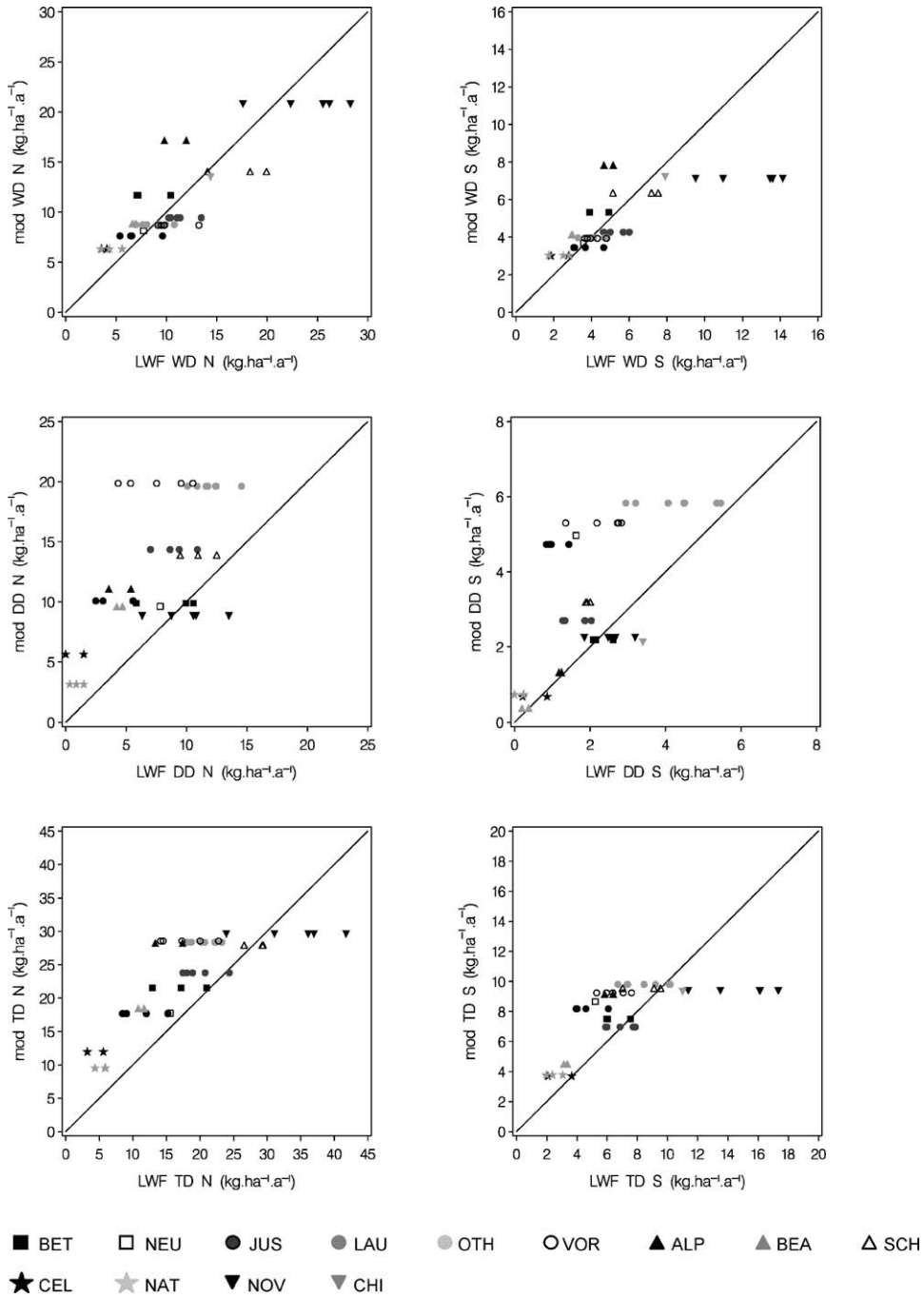


Figure 5. Modeled deposition versus LWF estimates. Left: N deposition (wet deposition WD, dry deposition DD, total deposition TD). Right: S deposition (WD, DD, TD).

TABLE VII

Empirical critical loads for nitrogen (CL<sub>N</sub>) and calculated critical loads using the steady state mass balance (SMB) method for the LWF plots

	$U_N$	$I_N$	$le(acc)_N$	fde	SMB CL <sub>N</sub>	Empirical CL <sub>N</sub>	TF <sub>N</sub>	TD <sub>N(LWF)</sub>	TD <sub>N(mod)</sub>
BET	0	4.3	3.1	0.3	8.8	15–20	13.6	17.0	21.6
NEU	0	3.2	3.9	0.3	8.7	15–20	10.8	15.6	17.8
JUS managed	7.1	3.0	4.0	0.3	15.9	10–20	9.2	11.2	17.7
JUS unmanaged	0	3.0	4.0	0.3	8.7	10–20	9.2	11.2	17.7
LAU	7.1	3.6	3.6	0.3	15.9	10–20	15.0	19.9	23.8
OTH	7.1	3.0	4.0	0.5	18.1	10–20	15.9	20.8	28.4
VOR	4.3	3.0	4.0	0.3	13.0	7–20	14.6	17.8	28.6
ALP	2.2	4.3	3.1	0.7	16.9	15–20	13.5	15.4	28.3
BEA	2.2	5.0	2.7	0.3	10.9	7–20	9.4	11.3	18.5
SCH	4.3	3.5	3.7	0.3	13.0	10–20	22.3	28.4	27.9
CEL	1.5	5.0	2.2	0.3	9.6	7–20	3.5	4.4	12.0
NAT	0	5.0	2.1	0.3	8.1	15–20	4.4	5.4	9.6
NOV	0	3.9	3.4	0.3	8.8	10–20	29.2	34.0	29.6
CHI	0.8	4.7	2.8	0.3	9.6	7–20	20.1		21.0

*Note.* The SMB method uses the following equation:  $CL_N = U_N + I_N + le(acc)_N/(1-fde)$  (in Rihm 1996) where  $U_N$  is the amount of  $N$  removed by wood harvesting, fde the denitrification fraction,  $le(acc)_N$  the acceptable total leaching from the rooting zone,  $I_N$  the acceptable immobilization rate of  $N$  in soil organic matter. All values (except fde) are in  $kg N ha^{-1} a^{-1}$ .

499 deposition levels would be in the absence of anthropogenic sources) and below the  
 500 critical loads for  $N$ . Similar low deposition rates for  $N$  were measured in the second  
 501 half of the 1980s in the Alps in Davos (Kloeti *et al.*, 1989), but the throughfall depo-  
 502 sition of  $S$  was higher in Davos ( $10 kg S ha^{-1} a^{-1}$  under a spruce canopy) than that  
 503 measured at CEL and NAT. Different stand characteristics, the vicinity of the town  
 504 of Davos, or a general decrease in  $S$  deposition might be possible explanations. At  
 505 CEL and NAT, TF fluxes were only slightly higher than WD fluxes (derived from  
 506 BP fluxes) for  $N$  and  $S$ , indicating a very low contribution of dry deposition to  
 507 the total deposition and/or canopy uptake of these elements. For  $N$  this pattern is  
 508 typical of remote areas (Lovett and Lindberg, 1993). At NAT the availability of  $N$   
 509 for vegetation appears to be low as the carbon nitrogen ratio (C:N) in the soil is very  
 510 high and the decomposition rate of organic matter slow (Dobbertin *et al.*, 2001).  
 511 At CEL, the C:N ratio in the soil organic matter is lower, but the C:N in the soil  
 512 solution is high (Pannatier, WSL, personal communication). Hence, atmospheric  
 513 nitrogen is likely to be readily taken up by the canopy on these plots.

514 On the Central Plateau, deposition of  $N$  and  $S$  was moderate. Mean annual  $SO_4^{2-}$   
 515 concentrations in WD at OTH and VOR were very close to those measured in 1997–  
 516 1999 at the EMEP stations on the Plateau (Dübendorf, Payerne, Tänikon) (NABEL,  
 517 2000). Likewise, similar  $SO_4^{2-}$  concentrations in WD were measured at BET and

the EMEP station of Chaumont (Jura). The LWF measurements of S deposition were substantially lower than the deposition rates measured on the Lägern, in the lowlands near Zurich at the end of the 1980s (Kloeti *et al.*, 1989), both in the open area (10 kg S ha<sup>-1</sup> a<sup>-1</sup> in WD on the Lägern) and in throughfall (15.1 kg ha<sup>-1</sup> a<sup>-1</sup> under beech canopy, 40.3 kg ha<sup>-1</sup> a<sup>-1</sup> under spruce canopy). In 2001, Burkard *et al.* (2003) estimated the wet deposition of sulfur on the Lägern to be about 4.2 kg ha<sup>-1</sup> a<sup>-1</sup>, which is within the range of the LWF values. This is in agreement with the decrease in the wet deposition of sulfur also observed at Dübendorf and Payerne over the period 1985–1999 (NABEL, 2000) and the general reduction in S emissions in Switzerland (FOEFL (Publisher), 2002).

In contrast, the N deposition in the open areas on the LWF plots on the Plateau differed little from the wet deposition on the Lägern in the 1980s (9.1 kg N ha<sup>-1</sup> a<sup>-1</sup>; Kloeti *et al.*, 1989), and was only slightly higher than in 2001 at this latter site (about 6.9 kg N ha<sup>-1</sup> a<sup>-1</sup>; Burkard *et al.*, 2003). Under a spruce canopy, throughfall deposition was substantially higher on the Lägern (35.7 kg N ha<sup>-1</sup> a<sup>-1</sup>) than on any of the LWF plots located on the Plateau, but the N throughfall fluxes under a beech canopy on the Lägern (19.3 kg N ha<sup>-1</sup> a<sup>-1</sup>) were only 1.3 kg N ha<sup>-1</sup> a<sup>-1</sup> higher than the highest TF flux at OTH (measured in 1999) in a similar stand. Air concentrations of nitrogen oxides decreased from the end of the 1980s to 1994 and appear to be stabilizing. There do not seem, however, to be such clear trends for NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> concentrations in wet deposition (NABEL, 2002). At OTH, the reduced form of nitrogen rather than the oxidized form seems to account for the higher N fluxes on this plot (Table IV, Figure 3), probably due to the fact that the OTH forest stand is near to agricultural fields and that ammonia tends to be readily re-deposited locally (e.g. Sutton *et al.*, 1998). The concentrations of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in WD at OTH, VOR and LAU were similar to those measured in 1997–1999 at EMEP stations on the Central Plateau, i.e. 35–45 mg l<sup>-1</sup> of NO<sub>3</sub><sup>-</sup>-N and 40–55 mg l<sup>-1</sup> of NH<sub>4</sub><sup>+</sup>-N (NABEL, 2000).

NOV and SCH, which had the highest deposition of nitrogen, were on the other end of the deposition range. At NOV, sulfur and proton depositions were also the highest, whereas the S deposition on all the other plots was low to moderate, reflecting the large decrease in SO<sub>2</sub> emissions during the last 15 years. N and S deposition in the open area of NOV was in the upper range of deposition measured at several stations in Southern Switzerland (Barbieri and Pozzi, 2001) and the northern part of Italy bordering Switzerland (Della Lucia *et al.*, 1996; Balestrini and Tagliaferri, 2001). The high deposition rates at NOV are related to the high level of emissions from the industrialized and densely populated Po Basin to which the southern part of Switzerland is subjected and the relatively high amount of precipitation compared to the open Po Plain (Della Lucia *et al.*, 1996). The Southern Alps have also been recognized as being most sensitive to acid deposition (Rihm, 1996). The high deposition rates at SCH, which clearly exceeded the depositions measured on the two other plots located in the Lower Alps (ALP, BEA), are most likely related to the agricultural activities (livestock farming) carried out in the plain

561 and to the geographic situation of the plot. It is located on the northern flank of the  
562 Alps, where an orographic barrier effect occurs.

#### 563 4.2. COMPARISON WITH MODELED DEPOSITION

564 For nitrogen, LWF BP measurements were close to the modeled WD. However,  
565 for all five plots north of the Alps above 1000 m (BET, ALP, BEA, CEL, NAT),  
566  $WD_{N(mod)}$  was higher than  $WD_{N(LWF)}$ . The difference appears to be related to the  
567 modeled N concentrations in precipitation, which were systematically higher than  
568 the corrected concentrations measured by LWF. This suggests that the parameters  
569 of the linear equation used to model N concentrations above 1000 m should be  
570 adjusted. At ALP, the modeled precipitation was moreover noticeably higher than  
571 the precipitation amounts measured by LWF over the two sampling years, due to  
572 the lower than average precipitation amounts at the site during these two years,  
573 which increased the difference between  $WD_{N(mod)}$  and  $WD_{N(LWF)}$ . At plots below  
574 1000 m, the concentrations measured by LWF were scattered around the modeled  
575 concentration, assumed to be constant ( $0.45 \text{ mg NH}_4\text{-N l}^{-1}$ ,  $0.33 \text{ mg NO}_3\text{-N}$   
576  $\text{l}^{-1}$ ), but  $WD_{N(mod)}$  and  $WD_{N(LWF)}$  were in close agreement except at SCH. For the  
577 Southern Alps,  $\text{NH}_4^+$  and  $\text{NO}_3^-$  concentrations in wet deposition resulting from the  
578 model of Barbieri and Pozzi (2001), which we applied to this part of Switzerland,  
579 were both lower than the measured concentrations. However, this might be related  
580 to the specific local characteristics of NOV, and possibly CHI as well, rather than to  
581 an inaccuracy of the model, which has been calibrated with recent data (1993–1998)  
582 from 13 sites in Southern Switzerland (Barbieri and Pozzi, 2001).

583 In contrast,  $DD_{N(mod)}$  was noticeably higher than  $DD_{N(LWF)}$  except at NOV. As  
584  $DD_{N(LWF)}$  is itself derived from model calculations, it is difficult to determine which  
585 of  $DD_{N(mod)}$  and  $DD_{N(LWF)}$  is closest to the true value for  $DD_N$ . In a detailed study of  
586 atmospheric deposition in a spruce stand in Alptal close to the ALP LWF plot,  $DD_N$   
587 estimated with inferential modeling amounted to approximately  $5 \text{ kg N ha}^{-1} \text{ a}^{-1}$   
588 (Fischer-Riedmann, 1995). This value is consistent with  $DD_{N(LWF)}$  ( $4.5 \text{ kg N ha}^{-1}$   
589  $\text{a}^{-1}$ , Table VI) whereas  $DD_{N(mod)}$  ( $11.1 \text{ kg N ha}^{-1} \text{ a}^{-1}$ ) is higher. Measurements of  
590  $\text{NH}_3$  and  $\text{NO}_2$  on selected LWF plots during 2000 using passive samplers should  
591 help to determine the causes of the discrepancies between the modeled and estimated  
592 dry deposition values by supplying measured concentrations which can be directly  
593 compared to the concentrations used in the resistance model.

594 It is also worth noticing that the inter-annual variability of LWF deposition mea-  
595 surements was high, especially at NOV, reflecting at least partly the high variability  
596 of precipitation. The comparison could yield an entirely different picture if only  
597 one sampling year was available. In 1999, when the measured deposition fluxes at  
598 NOV were highest ( $WD_{N(LWF)} = 28.3 \text{ kg ha}^{-1} \text{ a}^{-1}$ ,  $DD_{N(LWF)} = 13.5 \text{ kg ha}^{-1} \text{ a}^{-1}$ ),  
599 the modeled values  $WD_{N(mod)}$  and  $DD_{N(mod)}$  would have been  $-7.5$  and  $-4.7 \text{ kg}$   
600  $\text{ha}^{-1} \text{ a}^{-1}$ , respectively, lower than the LWF values. In contrast, in 1997, when the  
601 lowest LWF deposition fluxes were measured,  $WD_{N(mod)}$  and  $DD_{N(mod)}$  would have

exceeded LWF values by 3.2 and 2.5 kg ha<sup>-1</sup> a<sup>-1</sup>, respectively. Besides the precipita- 602  
tion amount, other factors could contribute to the large differences in deposition ob- 603  
served between years, for example, deposition from alkaline dust, transported from 604  
the Sahara, occurs occasionally and can result in peaks of base cations in the precipi- 605  
tation samples, which, in turn, influences the output of the canopy budget model. 606

With regard to sulfur,  $WD_{S(LWF)}$  and  $WD_{S(mod)}$  were in close agreement, except 607  
at NOV, and, to a lesser extent, ALP. In contrast,  $DD_{S(mod)}$  tended to be higher 608  
than  $DD_{S(LWF)}$ , even though the model was expected to underestimate sulfur dry 609  
deposition since it calculated SO<sub>2</sub> dry deposition only, ignoring particulate sulfate 610  
deposition. As a result of elevated  $DD_{S(mod)}$ ,  $TD_{S(mod)}$  also tended to be higher 611  
than  $TD_{S(LWF)}$ .  $DD_{S(mod)}$  exceeded  $DD_{S(LWF)}$ , except at NOV, on all the broadleaved 612  
plots (NEU, JUS, LAU, OTH, SCH) and at VOR. At VOR, *Abies alba* is the main 613  
species, but VOR is actually a mixed stand with a number of broadleaved species. 614  
This suggests that, instead of applying a single deposition velocity factor to the 615  
modeled SO<sub>2</sub> concentrations regardless of forest type, a distinction should be made 616  
between broadleaved and coniferous stands. 617

#### 4.3. EXCEEDANCE OF CRITICAL LOADS

618

Rihm (1996) reported that the critical loads for N in managed forests calculated with 619  
the SMB method were exceeded on 95% of the mapped area. Using updated data 620  
for the calculation of N deposition (i.e. reference years: 2000 north of the Alps and 621  
1993–2000 south of the Alps instead of 1993–1995 in Rihm's report, and a lower 622  
NH<sub>3</sub> deposition velocity for broadleaved stands), the SMB critical loads are still 623  
exceeded on 92% of the mapped forest area. The same overall picture is achieved 624  
if we compare  $TD_{N(mod)}$  to the SMB critical loads on the LWF plots: for all plots, 625  
critical loads for N are exceeded. The excess range was greater in Rihm's report than 626  
for the LWF plots, however, due to the different years of reference used to model 627  
N deposition and the higher deposition velocity applied to NH<sub>3</sub> concentrations in 628  
the 1996 report. In addition, the parameter defining the total acceptable N leaching 629  
in the equation calculating the critical loads was also modified. 630

If  $TD_{N(LWF)}$  is compared to the SMB critical loads, the overall picture shows more 631  
contrasts, with two plots below 1800 m as well as both plots above 1800 m receiving 632  
total N deposition lower than the calculated  $CL_N$ . When comparing  $TD_{N(LWF)}$  with 633  
the empirical  $CL_N$ , it is even more difficult to make conclusive statements since 634  
 $TD_{N(LWF)}$  was within the empirical  $CL_N$  but not above the upper value of the range 635  
for most plots. Only at NOV and SCH did atmospheric deposition of nitrogen 636  
exceed both modeled and empirical critical loads. 637

In addition, when comparing total deposition and calculated  $CL_N$ , it must be 638  
considered that a substantial margin of error is associated with the critical loads 639  
calculated according to the steady state mass balance (SMB  $CL_N$ ). The denitrifi- 640  
cation fraction,  $f_{de}$ , can only be a very rough estimate of the actual denitrification 641

642 rate. Moreover, the degree of hydromorphy derived from the soil suitability map  
643 did not always match the degree of hydromorphy indicated by features of oxidation  
644 reduction assessed on the soil profiles dug on the plots themselves (Walthert, WSL,  
645 personal communication; Walthert *et al.*, 2003). The maximum deviation, which  
646 can be expected from the “true” value, assuming an acceptable leaching of 4 kg N  
647  $\text{ha}^{-1} \text{a}^{-1}$  and an error on fde of 0.5, is as large as 8 kg N  $\text{ha}^{-1} \text{a}^{-1}$ . The value of  $U_N$   
648 (N removal by wood harvesting) is based on regional means, which could be quite  
649 different from the actual harvesting rates, so that deviations in the same order of  
650 magnitude are to be expected.

#### 651 4.4. UNCERTAINTIES IN FLUXES

652 Uncertainties are involved in both the throughfall method and the modeling ap-  
653 proach. In the model, one of the main uncertainties probably lies in the deposition  
654 velocities. The values of deposition velocities used in the model were either con-  
655 stant or depended on the type of stand (coniferous or broadleaved) or altitude only.  
656 We expect that the model results could be improved by taking into account local  
657 meteorological and canopy characteristics in the setting of the deposition veloci-  
658 ties. Air concentrations of  $\text{NH}_3$ ,  $\text{NO}_2$  and  $\text{SO}_2$ , which were modeled on the basis of  
659 the data from 40 to 120 monitoring stations, should be quite reliable. The modeled  
660 concentrations of aerosols and concentrations in precipitation, on the other hand,  
661 rely on a very limited set of monitoring data. Our measurements of bulk deposition  
662 indicate that a better calibration of the model, which calculates concentrations in  
663 precipitation, is needed. Since the calculated wet deposition at the plot level is a  
664 linear function of concentrations and precipitation amounts, a 50% variation in one  
665 of the input parameters would result in a 50% variation in the calculated wet depo-  
666 sition. Likewise, a 50% variation of the deposition velocity would result in a 50%  
667 variation of the dry deposition. Since dry deposition of N accounts for 30–70% of  
668 the total N deposition, a 50% variation in the deposition velocity would result in a  
669 variation of 15–35% in  $\text{TD}_N$ .

670 There are a number of uncertainties related to throughfall. The general uncer-  
671 tainties related to the method (e.g. the assumptions of the canopy budget model)  
672 should be kept in mind but are not dealt with here (EC-UN/ECE *et al.*, 2001).  
673 Several uncertainties are specifically related to LWF measurements. Some of them  
674 do not lead to systematic under- or overestimation of deposition measurements, in-  
675 cluding the use in winter of 1–4 snow collectors only on the LWF plots with difficult  
676 access in winter, while it is generally known that the spatial variation in throughfall  
677 fluxes at the plot scale can be substantial (see Thimonier (1998b) for an analysis of  
678 within-stand variability at OTH, and e.g. Houston *et al.* (2002)). Other factors bias  
679 estimates of atmospheric deposition in a systematic way, e.g. road salting: NaCl  
680 particles blown away by the wind could contaminate bulk precipitation collectors,  
681 which would decrease estimations of the dry deposition of base cations and nitro-  
682 gen in the canopy budget model. Also to be mentioned is a possible contamination

with  $\text{Na}^+$  and  $\text{Ca}^{2+}$  during sample preparation in the laboratory in 1999/2000, 683  
as suspected by occasionally higher values in blank samples during this period. 684  
Furthermore, since stemflow was not measured, atmospheric deposition was often 685  
underestimated, especially on the LWF stands where beech (*Fagus sylvatica* L.) 686  
is the main tree species (NEU, LAU, OTH, SCH). In beech stands, stemflow is 687  
substantial and can easily account for 10% of incident precipitation (in Thimonier, 688  
1998a). Its contribution to the total deposition can be even higher since it is usually 689  
more concentrated than throughfall (Parker, 1983). In a deciduous hardwood forest 690  
near Ithaca (NY, USA), Butler and Likens (1995) found that stemflow represents 691  
16% of the S deposition and 12–13% of the N deposition to the forest floor. 692

A field comparison of 20 different sets of sampling equipment and designs 693  
used within ICP Forests showed that precipitation amounts differed by 17.9% on 694  
average from measurements obtained with a collocated Hellmann gauge in an 695  
open area (Draaijers *et al.*, 2001). Precipitation measured by our collectors during 696  
this comparison was 13% higher than the precipitation measured by the Hellmann 697  
gauge, which led to above average fluxes. This may have been due to the connection 698  
between the funnel, bottle and the fine-meshed filter used before March 2001 not 699  
being tight. Yet, the bulk precipitation amounts measured by LWF at ALP were in 700  
close agreement (0.9% lower in 1997 and 2.7% lower 1998) with those measured 701  
using a heated tipping bucket of a collocated WSL weather station which is set 702  
according to WMO standards (Stähli, WSL, personal communication; description 703  
of installation see e.g. Keller *et al.* (1989); Burch *et al.* (1996)). Inorganic nitrogen 704  
fluxes in BP were  $13.6 \text{ kg N ha}^{-1}$  for the hydrology project (Stähli, WSL, personal 705  
communication) and  $13.9 \text{ kg N ha}^{-1}$  for LWF. Other sampling equipment used 706  
in the NITREX project (see e.g. Schleppei *et al.*, 1998) in a nearby area measured 707  
similar precipitation amounts to those in LWF, but recorded approximately  $2 \text{ kg N}$  708  
 $\text{ha}^{-1}$  less nitrogen than LWF (Schleppei, WSL, personal communication). However, 709  
S fluxes measured in the NITREX and LWF projects were similar. It is possible that 710  
the observed differences in N fluxes could be ascribed to the different exposure of 711  
the open areas to local emissions of nitrogen, since the open areas of the NITREX 712  
and LWF projects are 250 m apart. At ALP, rain collectors were in use for part 713  
of the year only, but overall the comparison results suggest that the filter did not 714  
greatly influence our assessments of annual fluxes. 715

A further source of uncertainty is that the chemical composition of the samples 716  
over the two-week collection period may change, especially in summer. Nitro- 717  
gen compounds are especially unstable (see e.g. literature review by Thimonier, 718  
1998a), so we have not discussed in detail the relative contributions of  $\text{NO}_3^-$  and 719  
 $\text{NH}_4^+$  to the total dissolved inorganic N deposition at the LWF plots. A decrease 720  
in  $\text{NH}_4^+$  concentrations over time has been frequently reported (Liechty and Mroz, 721  
1991; Ferm, 1993). This may be due to micro-organisms transforming  $\text{NH}_4^+$  into 722  
organic forms or to nitrification. We have only presented the concentrations and 723  
fluxes of inorganic nitrogen. The inclusion of the dissolved organic fraction would 724  
yield higher nitrogen fluxes. In 2001, most of the samples were analyzed with the 725

726 combustion method, and for a rough estimate, we corrected the total dissolved  
727 nitrogen concentrations for those few samples having been analyzed with the alka-  
728 line persulfate oxidation method. The flux of DON in throughfall was then between  
729 0.8 and 4.2 kg N ha<sup>-1</sup> (lowest fluxes in CEL, highest fluxes in VOR), which rep-  
730 resented 10.7% (OTH) to 26.1% (VOR) of the total dissolved nitrogen flux under  
731 the canopy. In the open area, DON fluxes in BP were usually <1 kg N ha<sup>-1</sup> (OTH:  
732 1.2 kg ha<sup>-1</sup>). Before 2001, the corrected DON fluxes were much higher both in  
733 BP (on average 3.1 kg N ha<sup>-1</sup> a<sup>-1</sup>, with a maximum of 7.4 kg ha<sup>-1</sup> in OTH in  
734 1996) and in TF (up to 11.6 kg N ha<sup>-1</sup> in 1999 in LAU, which represented 39.7%  
735 of total dissolved nitrogen). Therefore, if dissolved organic N is included in the  
736 total N deposition, some of the plots for which the inorganic N deposition is within  
737 the critical loads range (Table VII) would be ranked among the plots where critical  
738 loads are exceeded.

739

## 5. Conclusion

740 In this comparison of modeled and measured atmospheric deposition on 13 LWF  
741 plots, we found that the wet deposition of N and S derived from bulk deposition  
742 measurements were close to the modeled values, whereas estimates of the dry  
743 deposition of both N and S with the throughfall method using the canopy budget  
744 model were generally lower than the modeled values. In this comparison exercise,  
745 the high year-to-year variability of deposition shown by our measurements should  
746 be kept in mind. This high inter-annual variability stresses the need of having long  
747 enough measuring periods to assess atmospheric deposition on a site.

748 As regards wet deposition, the few differences observed between  $WD_{N(mod)}$   
749 and  $WD_{N(LWF)}$  suggest that the linear model used to calculate N concentrations in  
750 precipitation above 1000 m should be improved. However, modeled and measured  
751 values were in agreement on the whole.

752 The difference between LWF estimates and modeled values of dry deposition  
753 is probably partly due to the deposition velocities and the air concentrations used  
754 in the model. Passive sampling of NH<sub>3</sub> and NO<sub>2</sub> on selected LWF plots during  
755 2000 (analysis in process) will make it possible to check the relevance of the air  
756 concentrations used in the model. The values of deposition velocities could be ad-  
757 justed more finely by including information on turbulence in the boundary layer  
758 of the atmosphere (e.g. wind speed, surface roughness of topology and vegeta-  
759 tion), as well as information on factors influencing the surface resistance (e.g.  
760 humidity).

761 The results of the throughfall method could also be improved. Especially for  
762 beech stands, stemflow measurements should be included when measuring through-  
763 fall. The actual contribution of stemflow to deposition fluxes will soon be assessed  
764 at OTH and LAU, where stemflow is currently being sampled. Further improve-  
765 ments could be achieved by quantifying specifically for each site the correction

factors used to derive wet deposition from bulk deposition by running wet-only deposition collectors and bulk collectors in parallel for a limited time period. The weak acids should be better quantified, and the assumptions of the canopy budget model also need to be experimentally tested for our site conditions. In particular, the relative exchange efficiencies of  $H^+$ ,  $NO_3^-$  and  $NH_4^+$  should be determined for the main tree species on the plots. In addition, alternative approaches should also be explored to estimate the dry deposition of N, which is the component with the most uncertainties involved (e.g. micrometeorological methods, which include eddy correlation and gradient techniques, but which are technologically complex and expensive; wash studies using natural or artificial leaves as collectors).

In spite of the discrepancies between measured and modeled values, both methods resulted in total deposition estimates of N which exceeded the calculated critical loads for N and which were within or above the range of empirical critical loads for most of the plots. The two plots located above 1800 m were, however, exceptions (CEL, NAT). On the plots where critical loads are exceeded, ecological changes are expected to occur or may already be underway. Tree growth may be stimulated at some plots, but negative changes, such as a decrease in the species diversity of the ground vegetation, nutrient imbalances, or nitrate leaching could also take place. LWF will continue to study the various compartments of the ecosystems at the LWF plots to detect possible dysfunction or imbalances.

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## **Queries**

Q1. Au: Please update.

Q2. Au: Please update and furnish complete bibliographic details.