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Are there signs of acidification reversal in freshwaters of the low mountain ranges in Germany?

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Abstract

The reversal of freshwater acidification in the low mountain ranges of Germany is of public, political and scientific concern, because these regions are near natural ecosystems and function as an important drinking water supply. The aim of this study was to evaluate the status and trends of acidification reversal after two decades of reduced anthropogenic deposition in selected freshwaters of the low mountain ranges in the Harz, the Fichtelgebirge, the Bavarian Forest, the Spessart and the Black Forest. In response to decreased sulphate deposition, seven out of nine streams investigated had significantly decreasing sulphate concentrations (all trends were calculated with the Seasonal Kendall Test). The decrease in sulphate concentration was only minor, however, due to the release of previously stored soil sulphur. No increase was found in pH and acid neutralising capacity (defined by Reuss and Johnson, 1986). Aluminum concentrations in the streams did not decrease. Thus, no major acidification reversal can currently be noted in spite of two decades of decreased acid deposition. Nevertheless, the first signs of improvement in water quality were detected as there was a decrease in the level and frequency of extreme values of pH, acid neutralising capacity and aluminium concentrations in streams. With respect to nitrogen, no change was determined for either nitrate or ammonium concentrations in precipitation or stream water. Base cation fluxes indicate increasing net loss of base cations from all ecosystems investigated, which could be interpreted as an increase in soil acidification. The latter was due to a combination of continued high anion leaching and significant reduction of base cation deposition. No major improvement was noted in biological recovery, however, initial signs of recovery were detectable as there was re-occurrence of some single macroinvertebrate species which were formerly extinct. The results of this study have important implications for water authorities, forest managers and policy makers: the delay in acidification reversal suggests a need for ongoing intensive amelioration of waters, a careful selection of management tools to guarantee sustainable management of forests and the reduction of nitrogen deposition to prevent further acidification of soils and waters.

Keywords: freshwater, acidification reversal, drinking water supply, forested catchments, Germany

Introduction

Anthropogenic atmospheric deposition has been shown to influence the biogeochemistry of forest ecosystems and accelerate soil and water acidification (Johnson *et al.*, 1991; Ulrich and Sumner, 1991). High nitrogen (N) and/or sulphate (SO_4) deposition has resulted in N and SO_4 accumulation in forest soils, and acidification of surface waters. Furthermore, loss of base cations from forest soils can be accelerated by acid rain (Rhode *et al.*, 1995; BML, 1997; Yanai *et al.*, 1999). The decrease in anthropogenic deposition of protons (H^+) and SO_4 in German forest ecosystems during the last two decades has raised questions concerning the reversibility of acidification (defined here as reversing trends of chemical parameters) and the biological recovery of ecosystems.

Trends in freshwater acidification and the recovery of streams and groundwaters in the low mountain regions of Germany are of public, political and scientific concern, because the forested catchments of these regions are near natural ecosystems and are a major source of drinking water. The allowable range of acidity for drinking water in Germany is $6.5 \leq pH \leq 9.5$ (LfW, 1997). Even though acidified water in itself is not toxic to humans as long as pH is between 4 ≤ pH ≤ 10, acidified waters will accelerate plumbing corrosion and contribute to leaching of potentially toxic and/or irritating metals such as lead (Pb), iron (Fe), copper (Cu), aluminium (Al) and manganese (Mn) (LfW, 1997). Water acidity in forested catchments is connected to leaching of Al, Fe, and Mn from forest soils. High Al concentrations in waters have been shown to be toxic for fish (Driscoll *et al.*, 1980; Baker and Schofield, 1982), and amphipods (Musibono and Day, 1999) and are suggested to be harmful for humans (Struys-Ponsar *et al.*, 2000; Yokel, 2000). Human neurotoxicity of metals has been shown to be increased by interactions of various metal species such as Al, Fe, Mn, Cu and zinc (Zn) (Verity, 1999; Christen, 2000). Furthermore, the acidification of soils and soil solutions has been shown to cause problems with forest nutrition and is one cause for forest dieback (Lorenz, 1995; Müller-Edzards *et al.*, 1997; Alewell *et al.*, 2000a), even though the link between forest condition and soil acidification is difficult to validate (Cronan and Grigal, 1995; Erisman *et al.*, 1997; De Wit, 2000).

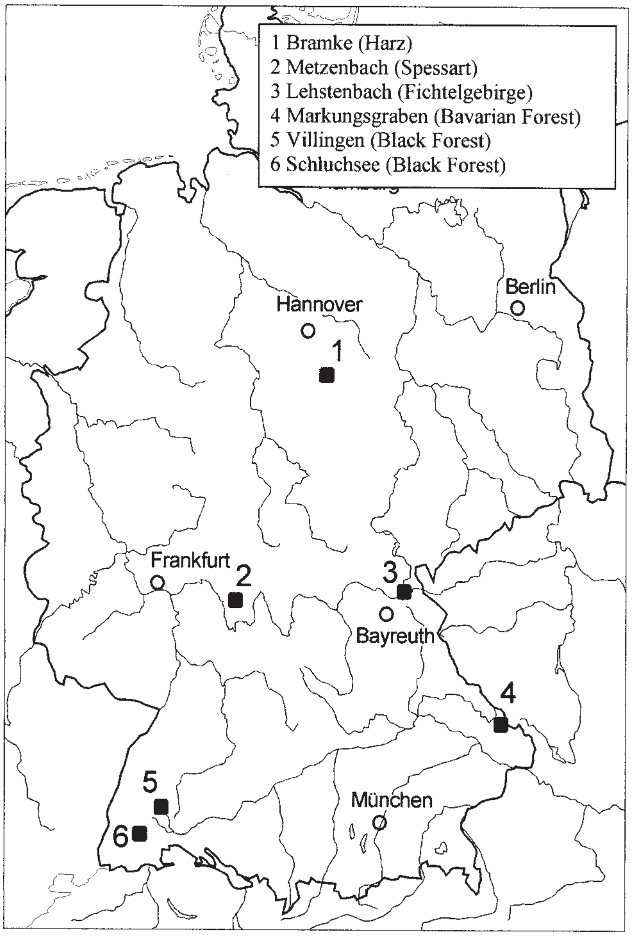

Fig. 1. *Location of sites*

Regions investigated in this study are low mountain ranges in Germany (see Fig. 1). Groundwater and springs draining these regions are an important drinking water resource in acidified regions in Bavaria. The major part has been shown to exceed the permitted drinking water contaminant level for several parameters between 1988 and 1994 (LfW, 1997), prompting additional efforts for water treatment (deacidification, Al purification) especially in small water plants. Of the freshwaters investigated (mostly untreated water samples from groundwater supplies) in the Spessart, the Fichtelgebirge and the Bavarian Forest, 87, 94, and 98%, respectively, had average pH ≤ 6.5 between 1988–1994 (n = 62, 74, 120, respectively, LfW, 1997). Approximately 20-40% of the groundwater samples had Al > 0.05 mg l^{-1}, which is the drinking water guideline value recommended by the EU (note that the drinking water contaminant level in Germany is 0.2 mg l^{-1} which is exceeded in 3, 19 and 9% of the investigated groundwaters in the Spessart, the Fichtelgebirge and the Bavarian Forest, respectively; LfW, 1997). A similar situation is found for the Black Forest where pH values < 5 and Al concentrations > 0.2 mg l^{-1} were frequently measured in groundwaters (Hinderer and Einsele, 1998). One consequence of freshwater acidification is an increasing effort to ameliorate water quality (mainly deacidification and Al purification) before consumption as drinking water. Between 1988 and 1994, 21% of all treated waters (average values for all low mountain ranges in Bavaria investigated by the Bavarian Water Management Agency) had pH ≤ 6.5 which is outside the contaminant level for drinking water (LfW, 1997). In the case of Al, 68% of all treated drinking water had values above 0.05 mg l^{-1} and 15% exceeded 0.2 mg l^{-1} between 1988 and 1994 (LfW, 1997).

The biological recovery of streams draining the low mountain ranges in Germany is of minor public concern because in many streams fish have been absent for several decades. Nevertheless, the biological recovery of species communities in these streams is of ecological value which should not be ignored. In some streams of the Harz Mountains (Lange Bramke) or the Black Forest trout are still present in spite of acidification (Gebhardt, 1994; Leßmann *et al.*, 1994).

The aim of this study was the evaluation of trends in freshwater acidification in the low mountain ranges of Germany, namely, the Spessart, the Fichtelgebirge, the Bavarian Forest, the Harz mountains and the Black Forest. The likelihood of an acidification reversal in the near future and the consequences for water authorities, forest management and policy makers are discussed.

Methods

SITE DESCRIPTIONS

The Bramke area is located in the Harz mountains. The Lange Bramke catchment (51°52'N, 10°26'E) with an altitudinal range of 543 to 700 m a.s.l. has an area of 76 ha and is stocked with 54 years old spruce. Lange Bramke spring is a subcatchment of Lange Bramke. Dicke Bramke and Steile Bramke with catchment areas of 32 and 38 ha, respectively, are stocked with 20 to 110 years old spruce stands. The Steile Bramke catchment was limed in 1989 with 16 t ha^{-1} of dolomitic limestone (Meesenburg *et al.*, 2001). Average annual precipitation (1949–1995) was 1230 mm and average temperature 6°C (Schmidt, 1997). Bedrock consists mainly of sandstones with layers of calcareous sand and clay schist (geological formation Oberemsstufe of the Lower Devonia). Soils comprise dystric Cambisols and haplic Podzols. A small proportion (4.5%) of the Lange Bramke catchment is permanently waterlogged. The upland soils are acidic with $2.8 < pH(CaCl_2) < 4.4$. Per cent base saturation of cation exchange capacity (CEC) is below 6% in the upper 20 cm and below 10% in the lower mineral soil (down to 1 m depth).

The Metzenbach catchment (240 ha, 49°54'N, 9°26'E) is located in the low mountain range, Spessart, southeast of Frankfurt at an elevation of 380–568 m a.s.l. Bedrock consists mostly of fine grained sandstones (geological formation Bunter Sandstone) and the soils are predominantly Cambisols. 20% of the catchment soils are influenced by permanent or temporary water saturation. Soils are very acidic with $pH(CaCl_2)$ between 2.9 and 4.0. Base saturation is below 15% in the upper soils but increases to >20% in 2–4 m depth. Mean air annual temperature is 7°C and annual average precipitation is 950–1100 mm (1988–1999). The catchment is stocked with beech and oak.

The Lehstenbach catchment (4.2 km^2, 50°09'N, 11°52'E) is located in the Fichtelgebirge area in Northern Bavaria close to the border with the Czech Republic at a height of 695–875 m a.s.l. The granite bedrock was deeply weathered during the Tertiary. The average precipitation in the area is 1000 mm and the annual average temperature is 6°C (1988–1999). The soils in the upland part of the catchment are acidic and have been classified as dystric Cambisols and Podzols according to FAO-classification. 30% of the catchment is covered by boggy areas where soils have been classified as fibric Histosols and dystric Gleysols. Base saturation of the mineral soils down to 2.5 m is less then 10%. Ninety per cent of the catchment is stocked with Norway spruce (*Picea abies* (L.) Karst.) of various age classes.

The Markungsgraben catchment (48°57'N, 13°25'E) is a subcatchment of the catchment Große Ohe in the Bavarian Forest National Park on the Bavarian-Czech border. Markungsgraben has an area of 1.1 km^2 at an altitude of 890–1355 m a.s.l. Average slope inclination is 27%. Bedrock consists of coarse granite and gneiss with deeply weathered portions and periglacial and glacial overburden (Moritz and Bittersohl, 2000). Mineral soils (classified as Cambisols and dystric Cambisols) are acidic ($pH(CaCl_2)$ 3.5 to 4.0) and base saturation is below 10% at 30 cm depth. The catchment is completely forested with spruce in the highland region and mixed woodlands in the lower slope regions. Average annual precipitation is 1680 mm with an average annual air temperature of 5.6°C (1989–1996, Moritz and Bittersohl, 2000).

The Villingen (46 ha, 48°03'N, 8°22'E) and Schluchsee (11 ha, 47°49'N, 8°06'E) catchments are both located in the Black Forest at altitudes of 870 – 950 m a.s.l. and 1150–1250 m a.s.l., respectively. Both catchments are stocked with spruce with an average age of 110 and 55 years, for Villingen and Schluchsee, respectively. Bedrock is sandstone (geological formation Bunter sandstone) in the case of Villingen and granite (geological formation Bärhaldegranit) in Schluchsee. Soils in Villingen are acidic and have developed to dystric Cambisols (70%) and dystric Planosols (30%) (Armbruster, 1998). Soil $pH(CaCl_2)$ is < 4 and base saturation < 7% down to 1 m depth, with the exception of a clay layer at 70 cm depth in the Planosols with a base saturation of 18%. Average air temperature at Villingen is 6.3°C with an annual bulk precipitation of 1330 mm (1988–1995) (Armbruster, 1998). Soils in the catchment Schluchsee are dominated by haplic Podzols with extremely low base saturation (< 4.1%) and $pH(CaCl_2) < 4.3$ down to 1 m depth. Average annual precipitation at Schluchsee between 1988 and 1996 was 1870 mm with an average air temperature of 4.4°C (Armbruster, 1998).

Average total deposition at the sites investigated is presented in Table 1 (for calculation of total deposition see below).

Table 1. Average total deposition of S and N in $mmol_c$ m^{-2} yr^{-1} at the catchments (arithmetic means of 1988–1995).

Catchment	*SO_4*	*NH_4*	*NO_3*
Lange Bramke	183	112	97
Lehstenbach	223	106	76
Markungsgraben	128	92	75
Metzenbach	67	55	46
Villingen	59	54	38
Schluchsee	58	44	37

A detailed site description as well as a description of sampling technique and analytical methods is given by Manderscheid and Göttlein (1995) for the Lehstenbach catchment (Fichtelgebirge), LfW (1994) for the catchments Metzenbach and Markungsgraben, by Armbruster (1998) for Villingen and Schluchsee (Black Forest) and by Schmidt (1997) for the Bramke area (Lange Bramke, Steile Bramke and Dicke Bramke catchments in the Harz mountains). Note that within the Bramke area, deposition and throughfall measurements are carried out only in the Lange Bramke catchment.

Most time series started in 1987/1988 and ended in 1999 with the exception of the sites within the Bramke area in the Harz mountains (measurement period 1975–1999 at Lange Bramke and Dicke Bramke; 1983–1999 at Lange Bramke spring and 1987–1999 at Steile Bramke) and the two sites Villingen and Schluchsee (Black Forest, measurement period 1987–1995/1996). Measurements of concentrations in bulk precipitation, throughfall and run-off were carried out weekly (Villingen, Schluchsee, note that starting in 1990 weekly precipitation and throughfall samples were combined to monthly bulk samples before analysis), fortnightly (Lehstenbach, Markungsgraben, Metzenbach) or monthly (Bramke area, weekly samples, monthly analyses).

Trend analysis was undertaken with the Seasonal Kendall Test, a non-parametric method in which data are grouped into seasonal (monthly) blocks, and ranked to identify monotonic long-term trends. The significance limit was set to $p < 0.05$, and trend slopes were estimated as the median concentration change between years for all seasonal blocks. The trend statistic T gives decrease (negative values) or increase (positive values) in $\mu mol_c l^{-1} yr^{-1}$. A detailed description of statistical methods is given in Evans *et al*. (2001).

CALCULATION OF FLUXES AND BUDGETS

Input fluxes with bulk precipitation were calculated as concentration times precipitation amount. Total deposition for all elements with the exception of SO_4 and N was calculated according to Eqn. (1) (Ulrich, 1994):

$$TD_{element} = ID_{Na} / BP_{Na} \; * BP_{element} \qquad (1)$$

with TD = flux of total deposition, ID = interception deposition ($ID_{Na} = TF_{Na} - BP_{Na}$), BP = flux of bulk precipitation and TF = flux of throughfall.

Several authors argue that SO_4 deposition with throughfall describes adequately patterns of total deposition, because of the low exchange between the forest canopy and S species (Matzner, 1988; Ulrich, 1994; Lovett *et al*., 1997). Thus, for total deposition of SO_4, throughfall flux values were taken. For all other ions, Eqn. (1) was used for the calculation of total deposition.

Output fluxes from the catchment with run-off were calculated according to:

$$\text{yearly element flux} = (\textstyle\sum \text{element flux at measurement day}) \; * \text{yearly discharge} / \textstyle\sum \text{discharge rate at measurement days}$$

Data of yearly output fluxes in Villingen and Schluchsee were taken from Armbruster (1998) who calculated fluxes according to Likens *et al*. (1977), where the sum of water fluxes of a measurement period were multiplied with the arithmetic means of element concentrations at the start and end point of the measurement period ("period-weighted-sample" method). Yearly element fluxes were calculated by summation of fluxes of all measurement periods. Comparative studies by Brahmer (1990) showed good agreements with fluxes calculated from weekly, time-proportional bulk samples.

For the calculation of budgets, output fluxes were subtracted from input fluxes. Thus, positive budgets indicate accumulation while negative budgets indicate net loss from the catchments.

When evaluating element fluxes in forested catchments, the quantification of water fluxes is crucial. At Markungsgraben and Schluchsee measurement of precipitation amount is connected to high errors because of steep slopes and relatively high contribution of snow to total annual precipitation (up to 40%). Thus, element fluxes in total deposition are most likely to be underestimated in these catchments (see Armbruster, 1998). At Metzenbach and Villingen part of the run-off leaves the catchment below ground. Armbruster (1998) measured and calculated the contribution of subterranean element fluxes via water supply wells to total catchment output for Villingen which is considered in the output fluxes presented. Subterranean element fluxes were calculated from measured groundwater concentrations and water fluxes obtained from two water supply wells inside the catchment. Subterranean water flux contributed 15% to the total catchment output, base cations between 17%–25%, H and Al 7 and 6%, respectively and SO_4, chloride (Cl) and NO_3 23, 16 and 25%, respectively (average values for 1988–1995) (Armbruster, 1998).

METHODS TO EVALUATE TRENDS IN BIOLOGICAL RECOVERY

To evaluate the status and trends in biological recovery the Bavarian Water Management Agency monitored macroinvertebrates and diatom communities between 1989 and 1996 in the Fichtelgebirge, the Spessart and the Bavarian Forest (investigated streams Eger, Speckkahl, Große Ohe, respectively; LfW, 1999). In the Harz mountains the stream Dicke Bramke was monitored for macroinvertebrates and diatom communities between 1988 and 1994 (LfW, 1998). With the help of macroinvertebrate communities the streams were classified in four categories of acidity as SZKL I to IV (SZKL = Säurezustandsklassen = acidity classes), which refer to not acid, low acidity, periodically acid and permanently acid. A detailed description of the sampling routine, calculation of acidity categories and evaluation of trends is given in LfW (1999).

Results and discussion

TRENDS IN CONCENTRATIONS

When discussing reversibility of acidification, SO_4 dynamics play a crucial role because SO_4 is the dominant anion in many soil solutions and waters. Sulphate leaching from soils causes cation leaching and is thus a key process in the acidification of soils and waters. Trends were calculated for total and estimated non-marine SO_4. Because of the relatively large distance from the sea, contribution of sea salt is very low in all catchments. Thus, trends for estimated non-marine SO_4 and total SO_4 are very similar. Because estimation of non-marine SO_4 is always connected to an unknown error (especially in areas with high dust emissions from industry), concentrations of total SO_4 are presented and discussed. Within the German catchments there was generally a significant decrease in SO_4 concentrations in throughfall and bulk precipitation (Table 2). Rather small changes in the SO_4 concentrations in throughfall and bulk precipitation where observed, however, in the Villingen and Schluchsee catchments in the Black Forest, an area that receives low deposition input (Table 1). In the case of throughfall, this change was significant ($p < 0.053$; Table 2) even though extreme values and peak frequency are clearly decreasing (data not shown, see Prechtel *et al*., 2001; Armbruster, 1998).

Table 2. Trends of statistics from the Kendall tau test in $\mu mol_c l^{-1} yr^{-1}$ for throughfall (TF) and run-off (Run) concentrations (significance level $p < 0.05$). Trends of bulk precipitation are given in brackets if significantly different from TF. NS = not significant. Trends for base cations were calculated as (∑ Ca, Mg, K, Na). ANC = calculated as Reuss and Johnson (1986). Note that NH_4 in runoff is mainly below detection limit.

	Base Cations		*Al*	*pH*		*ANC*		*Sulphate*		*NH_4*		*NO_3*	
Catchment	*TF*	*Run*	*Run*	*TF*	*Run*	*TF*	*Run*	*TF*	*Run*	*TF*	*Run*	*TF*	*Run*
Villingen	NS	NS	NS	+0.06	NS	NS	NS	NS (–1.6)	–3.6	NS	NS	NS	+0.2
Schluchsee	NS (–1.4)	NS	NS	NS (–0.3)	NS	+3.0	NS	NS (–1.4)	-1.1	NS (–1.4)	NS	NS	NS
Lange Bramke	–18.2	NS[1]	-	+0.05	NS	+6.1	NS	–9.9	+1.2	NS	-	NS	NS[2]
Lange Bramke spring	-	–3.3	–17.5	-	NS	-	NS	-	–2.2	-	-	-	–3.1
Dicke Bramke	-	–10.6	–2.7	-	NS	-	–3.2	-	–4.8	-	-	-	NS
Steile Bramke	-	–25.6	-	-	NS	-	NS	-	–6.7	-	-	-	–9.2
Markungsgraben	–13.8	–NS[3]	NS[3]	+0.07	NS[3]	NS	NS[3]	–13.7	NS[3]	–2.3	NS[3]	–5.4	NS[3]
Metzenbach	–12.3	–4.6	NS	NS (+0.04)	0.06	NS	NS	–4.4	–2.7	NS	0.1	NS	–1.7
Lehstenbach	–20.8	NS	NS	0.07	NS	-	NS	–34.0	–5.2	–4.5 (NS)	NS	NS	NS

[1] break in Jan-89 before: +2.1, after –8.4.
[2] break in Jan-84 before: +3.1, after –1.7
[3] stream trends *Markungsgraben* given up to Jan-96

In response to decreased deposition levels, seven out of the nine streams investigated had significant decreasing SO_4 concentrations (Table 2). Sulphate was still the dominant anion, however, in the streams of all investigated catchments (average % SO_4 of $\sum\{Cl, NO_3, SO_4\}$ = 37% in Markungsgraben, 53% in Lehstenbach, 60% in Schluchsee, 66–70% in the streams of the Bramke area, 70% in Metzenbach and 75% in Villingen). Thus, even though SO_4 deposition has decreased significantly for the last two decades, the overall decrease in SO_4 concentrations in streams of the lower mountain ranges in Germany was minor. This delayed response was caused by a release of previously stored inorganic and organic soil S (for a detailed discussion of S dynamics and trends see Prechtel *et al*., 2001).

In addition to decreasing SO_4 deposition, trends in pH and acid neutralising capacity (ANC) in bulk precipitation and throughfall were increasing slightly or remained unchanged (Table 2). Streams of the catchments investigated were acid and had pH ≤ 6.5 most of the year with no temporal trend (Fig. 2). The reason for the constant acidity in the streams and the delayed increase of pH can be attributed to the buffering capacity of the soils. All soils investigated have received considerable loads of acidity in the past (Table 1) and the stored SO_4 and acidity will be released under the currently low deposition regime.

Average ANC in streams (ANC as defined by Reuss and Johnson, 1986; levels calculated as arithmetic mean values) ranged from approximately 75 $\mu mol_c l^{-1}$ at Lange Bramke, Dicke Bramke and Metzenbach, 33 and 15 $\mu mol_c l^{-1}$ at Villingen and Schluchsee, respectively, around 0 $\mu mol_c l^{-1}$ at Markungsgraben, –50 $\mu mol_c l^{-1}$ at Lehstenbach to an average of –140 $\mu mol_c l^{-1}$ at Lange Bramke spring (Fig. 3, note that of the Bramke area only Lange Bramke is shown). At Steile Bramke, which was limed in 1989, average ANC was around 100 $\mu mol_c l^{-1}$ before liming (1987 to 1989) and 180 $\mu mol_c l^{-1}$ thereafter (1989–1999, data not shown). According to the Kendall tau test statistics, none of the streams had significantly increasing ANC (Table 2, Fig. 3). The stream Dicke Bramke within the Bramke area (Harz)

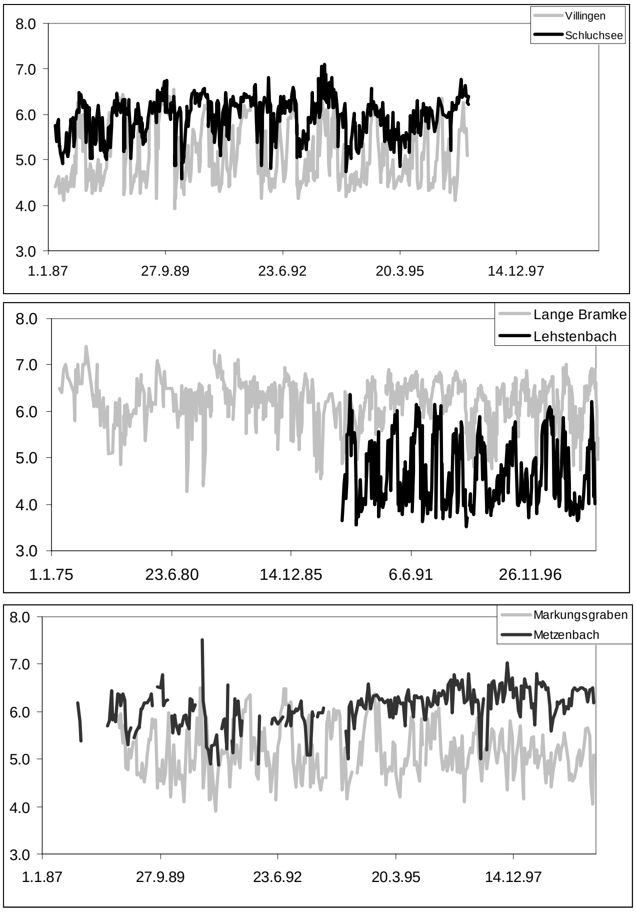

Fig. 2. *pH in stream waters*

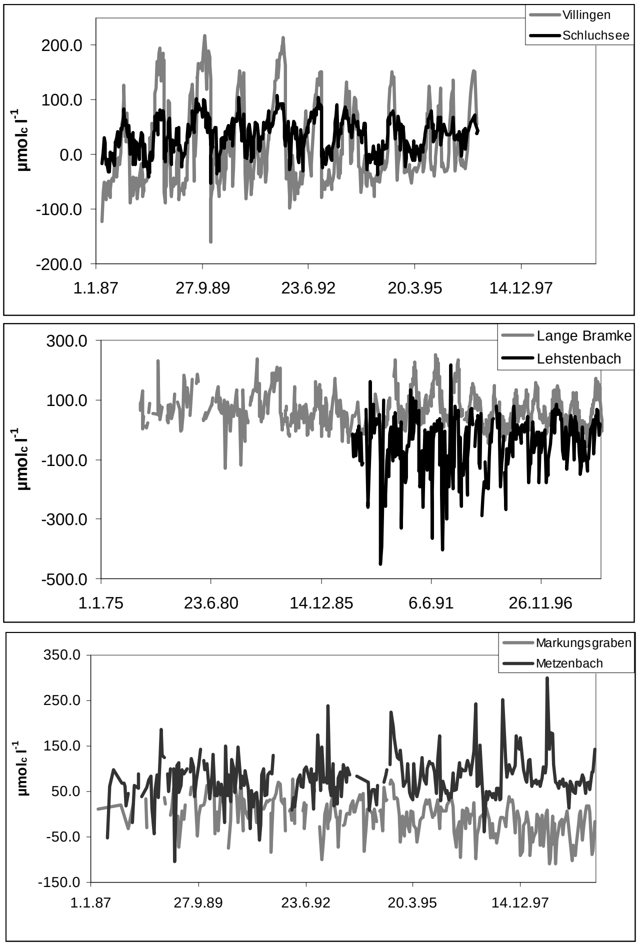

Fig. 3. *Acid neutralising capacity (ANC) of streams as defined by Reuss and Johnson (1986) in* $\mu mol_c l^{-1}$

even had significantly decreasing ANC (Table 2). Total Al concentration in streams responded similarly to pH and ANC. Kendall tau trend statistics (Table 2) and average stream concentrations of Al (Fig. 4) indicated no decline except for Lange Bramke spring and Dicke Bramke.

Although trends are not significant, a decrease in concentrations of extreme values and peak frequency is noticeable (Figs. 2, 3 and 4). The decrease in extreme values (both concentrations and frequencies) is clearly an indication of improved water quality and life conditions for the aquatic fauna. Thus, even though the decrease in acid deposition is so far not reflected in a major acidification reversal, the first signs of improvement are detectable.

With a reduction in particulate emissions throughout Europe and North America, base cation deposition has been reduced in many forest ecosystems (Driscoll *et al*., 1989; Hedin *et al*., 1994; Meesenburg *et al*., 1995; Alewell *et al*., 2000a;b). This is clearly reflected in the throughfall and bulk deposition trends of the investigated sites. At most sites the base cation concentrations in bulk precipitation and throughfall have decreased significantly (with the exception of the Villingen and Schluchsee sites in the Black Forest, Table 2). Parallel to the decrease in base cation concentration in the input to the catchments, four out of nine streams responded with a significant decline in base cation concentrations of stream water. At Steile Bramke, a decrease in base cation concentration in the stream occurred even though the catchment was limed. A general assessment of change in base cation concentrations in streams is not easy to make. A decrease in base cation concentrations in streams causes a delay in the increase of ANC (because ANC is calculated as the sum of base cations minus the sum of acid anions). On the other hand, continued high stream concentrations with decreasing input concentrations point to a continued high base cation leaching from soils (for a discussion of fluxes see below).

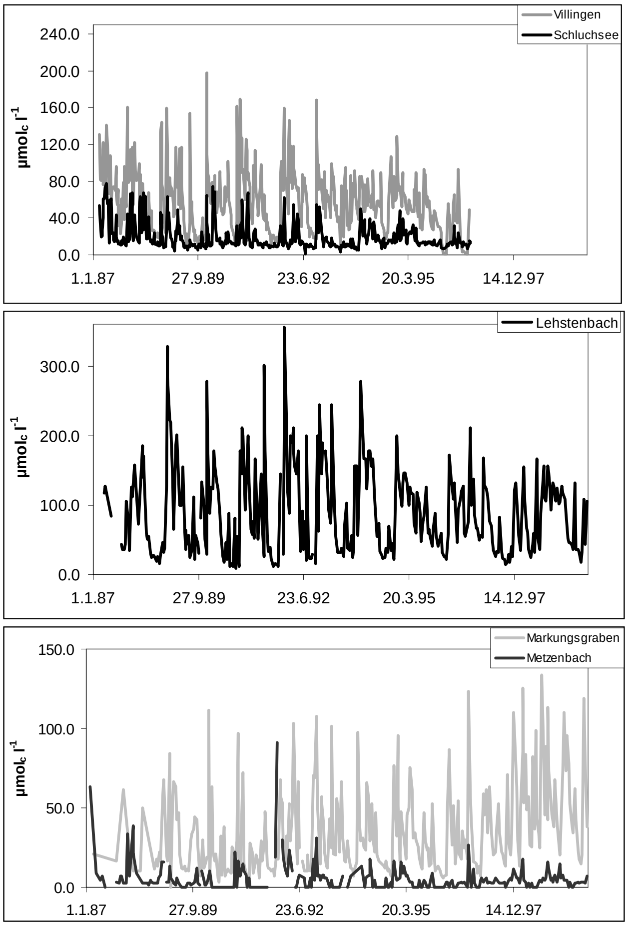

Fig. 4. *Aluminum concentrations in stream waters in µmol*$_c$*l*$^{-1}$

Inorganic N plays an important role in the acidification of soil solution and stream waters, and has often been underestimated in the past because of the capability of most terrestrial ecosystems to retain deposited N (Sullivan *et al*., 1997). A major part of the deposited NH_4 will be nitrified in acid forest soils, thus producing acidity within the soil (2 moles H per mole NH_4). Furthermore, leaching of Al, H and nutrient cations has been shown to be correlated to N saturation of soils (Currie *et al*., 1999). With the exception of Markungsgraben (Bavarian Forest) none of the sites decreased in either NH_4 or NO_3 concentration in throughfall and bulk precipitation (Table 2, note that at Markungsgraben bark beetle infestation caused dieback of the spruce stands in 1997 resulting in a decrease of interception deposition). Correspondingly, there are no clear trends in the NO_3 concentration of the streams. NO_3 concentrations decreased slightly in streams in Steile Bramke, Lange Bramke spring and Metzenbach . On average, over the measurement period, NO_3 contributed to 3% of "acid anions" (defined as $\sum$ Cl, NO_3, SO_4) in Villingen, 10% in Lehstenbach and Metzenbach, 10–16% in the streams of the Bramke area, 25% in Schluchsee and 53% in Markungsgraben (average values, no trends were noticeable with the exception of Markungsgraben as discussed below). Even though SO_4 is still the dominant anion in the streams of all catchments, NO_3 will become more and more important with an expected acidification reversal due to reduction of SO_4 deposition (for a discussion of the overall N dynamics on a European scale see Wright *et al*., 2001).

A special situation occurs in the Markungsgraben catchment. The catchment is located within the Bavarian Forest National Park which has the management aim of "leaving nature alone". Starting in 1996, mass propagation of a bark beetle (*Ips typographus* L.) resulted in the total destruction of the spruce stands, which covered 81% of the catchment area in 1999 (for details see Zimmermann *et al*., 2000). The bark beetle infestation caused a reduction in N input (due to deforestation and thus lower interception deposition) and a dramatic increase in N leaching (for budgets see below). Statistical analysis indicated a break in trend calculations in 1996 with a decline in ANC (Kendal tau statistics for trends from Jan-96 and onwards T = –3.6 $\mu mol_c l^{-1} yr^{-1}$; not signifcant) and a sharp increase in NO_3 (T = 30 $\mu mol_c l^{-1} yr^{-1}$; $p < 0.052$), and Al (T = 69 $\mu mol_c l^{-1} yr^{-1}$, not significant). Note, that trends were not significant due to the relatively short measurement period after the infestation (1996–1999). The example of Markungsgraben demonstrates that ecosystems change relatively quickly from functioning as sinks for N to extreme sources when environmental changes or disturbances occur. In Markungsgraben, however, NO_3 was already the dominant anion in run-off before the bark beetle infestation (approximately 50% of $\sum$ Cl, NO_3, SO_4 up to 1995 and increasing to 75% in 1998).

TRENDS IN FLUXES AND BUDGETS

A common tool for assessing the accuracy of element budgets in ecosystems is the evaluation of Cl budgets, because Cl is considered to be relatively inert to internal cycling within the ecosystems. Element budgets for Cl (Fig. 5) are generally well balanced with the exception of a negative budget in Lehstenbach (stream water output > input with total deposition) which was due to high additions of road salt on the roads during winter periods (Lehmann, 2000).

Under the decreasing SO_4 deposition regime of the last two decades, catchments have changed from a net retention to a net loss of SO_4 since the beginning of the 1990s (Fig. 5). Thus, with the decrease in deposition, previously stored SO_4 is currently released from the soils into the streams. Since the leaching of anions is always connected to cation leaching, the release of SO_4 delays a reversal of freshwater acidification due to leaching of H, Al, Fe, and Mn (for a detailed discussion of S dynamics and its influence on freshwater acidification see Prechtel *et al*., 2001).

With regard to N budgets, no trends were observed for NH_4 or NO_3. With the exception of Markungsgraben, none of the catchments showed a net loss of N from the catchment. At Markungsgraben, average retention of total N was 70 $mmol_c m^{-2} yr^{-1}$ until 1997 (average NO_3 loss until 1996 = 40 $mmol_c m^{-2} yr^{-1}$). In 1998, a sudden net loss of total N (180 $mmol_c m^{-2} yr^{-1}$ with a NO_3 leaching of 270 $mmol_c m^{-2} yr^{-1}$) was induced by mass propagation of bark beetles.

When discussing the development of acidification reversal, the dynamic of base cations is of major importance because of the effect on ANC. Furthermore, a significant leaching of base cations will lead to a decrease in base saturation of soils and may cause problems with forest nutrition (Malmer, 1976; Ulrich *et al*., 1980; Baes and McLaughlin, 1984; BML, 1997; Riek and Wolff, 1998). Despite the significant decrease in acid deposition, leaching of base cations did not decrease. All catchments investigated showed continued high cation leaching, and a general increase in net release from the ecosystems is observed (Fig. 6).

A similar dynamic has been shown for other sites in Germany (the Solling, Wesselink *et al*., 1995), Norway (Birkenes, Christophersen *et al*., 1990) and the United States (Hubbard Brook Experimental Forest, Driscoll *et al*., 1989). Thus, even though base cation concentrations in streams have been declining, net losses from ecosystems are increasing due to the combination of significant decrease in base cation deposition and continued high leaching of anions. A gradual loss of base cations from soils is a natural feature of weathering and pedogenesis. Loss of base cations, however, can be accelerated by acid rain, by forest regrowth following harvest removals and by declining inputs of base cations from atmospheric deposition (Johnson *et al*., 1991; Yanai *et al*., 1999). Thus, an increased loss in base cations has important implications for forest managers, water authorities and policy makers. Low harvesting intensity and careful selection of tree species can lead to tighter element cycles, thus decreasing acidification and leaching of base cations (Kreutzer, 1994). Lenz *et al*. (1994) pointed out that the harvest removal in the Fichtelgebirge mountains led to an acid load within the soil comparable to the anthropogenic acid deposition. Additionally, liming or amelioration of soils can improve base saturation. In many German forest ecosystems soils have to be improved in order to carry out regeneration programmes successfully or to rebuild more natural forest ecosystems (Raben *et al*., 2000). Since all catchments investigated are representative regions for drinking water supply, water authorities have to calculate the treatment of waters, namely deacidification and purification from Al, Fe and Mn probably for decades to come.

Because of the high errors connected with the calculations, it might be argued that budget calculations for whole catchments are insufficient evidence to prove that pools of soil base cations are decreasing. Trends in element soil pools, however, especially in base cations, are difficult to detect from direct soil analysis due to constraints regarding spatial heterogeneity, sampling techniques (e.g. resampling of the same spots and horizons, retaining the same methods over time) and slow development of temporal trends (Yanai *et al*., 1999). A decrease in soil base cation pools has been

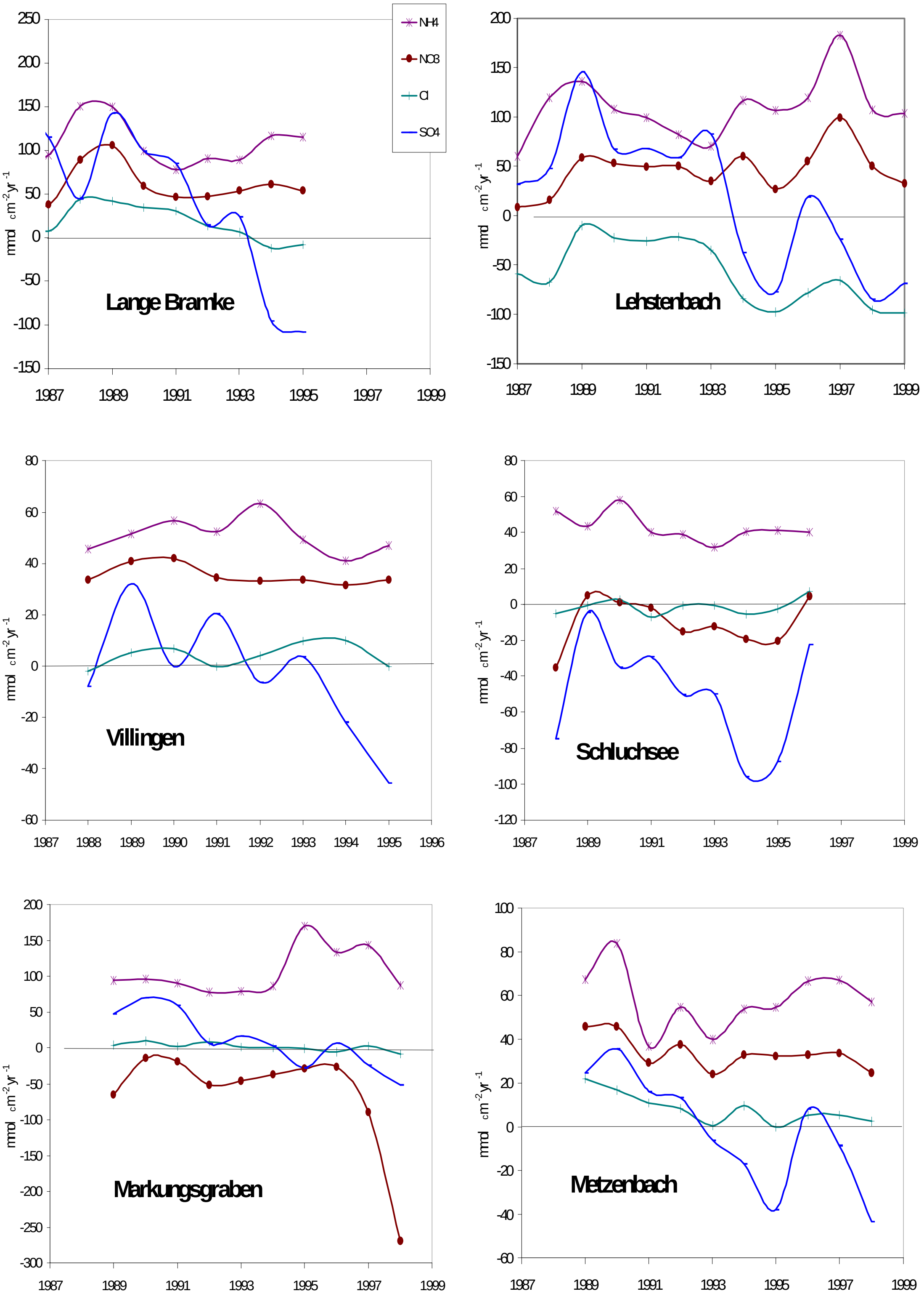

Fig. 5. *Element budgets (mmol$_c$m^{-2}yr^{-1}) for NH$_4$, NO$_3$, Cl and SO$_4$ calculated as total deposition minus run-off flux*

shown by Wesselink *et al*. (1995) for the Solling soil in Germany. Nevertheless, constraints regarding flux/budget calculations should not be ignored. Thus, conclusions from budget calculations in this study are based on trends in budgets (not absolute numbers) and only if supported by trends in concentrations.

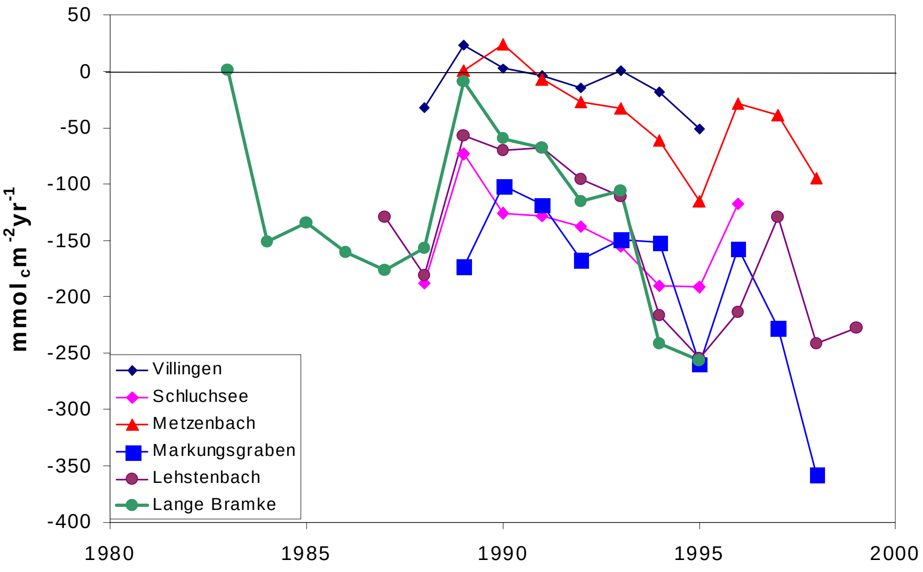

Fig. 6. *Element budgets for sum of base cations (Σ Ca, Mg, Na, K) in mmol$_c$m^{-2}yr^{-1} calculated as total deposition minus run-off flux.*

BIOLOGICAL RECOVERY

To judge the biological recovery in the Bavarian Forest data from the Große Ohe were assessed (note that Markungsgraben is a subcatchment of Große Ohe). Macroinvertebrates were classified according to the acidity category II - III (= between low acidity to periodically acid) with no clear trend between 1983 and 1996 (LfW, 1999). No acid sensitive species were found in the region, however, species from the genus *Baetis*, which are sensitive to acidity and were missing during the 1980s, were re-occurring in the early 1990s. Considering the development of the diatom communities, a decline in the abundance of *Eunotia exigua*, which is known to be acid tolerant, may be interpreted as a slight biological recovery (LfW, 1999). Biological recovery in three *Bavarian Forest* lakes (Schaumburg, 2000) was not observed, however, even though lake water chemistry showed signs of an acidification reversal (decrease of SO_4, Al, and NO_3 concentrations and increased pH).

Within the Fichtelgebirge area the stream Eger was investigated for trends in biological communities (Lehstenbach is a subcatchment of the Eger catchment). Macroinvertebrate communities improved from being between SZKL III to IV in the late 80s to SZKL III in the early 90s. Diatom communities, however, showed no trend. LfW (1999) pointed out, that the Eger stream drains an area which is protected from extremes of acid deposition by the mountains Schneeberg and Ochsenkopf, which function as a rain shield and so promote lower total deposition to the area. Thus, the slight improvement in acid status determined in this catchment is in contrast to other catchments investigated within the Fichtelgebirge mountains, most of which still show a general deterioration in biological status (LfW, 1999).

The stream Speckkahl in the *Spessart* mountains was investigated between 1994 and 1996. Macroinvertebrate and diatom communities point to an acidity category between low acidity to periodically acid (SZKL II – III; LfW, 1999) with no trends.

At Dicke Bramke, macroinvertebrate communities fluctuated between SZKL II and IV between 1988 and 1994. The occurence of *Helodes sp.* and *Odontocerum ablicorne* in the 1990s may indicate a beginning of biological recovery. Diatom communities indicated low acidity status with no trend throughout the measurement period (LWF, 1998).

Conclusions

Stream SO_4 concentrations in the low mountain ranges of Germany are generally decreasing due to reduced anthropogenic atmospheric SO_4 deposition. This is so far not reflected in an acidification reversal, as indicated by continuing low ANC and low pH of stream waters. First signs of reversibility of acidification in freshwater systems are indicated by decrease in concentrations and frequency of extreme values in ANC, pH, and Al. Net loss of base cations from the ecosystems is increasing due to decreased base cation deposition with continued high cation leaching. An increase of base cation loss from soils might be interpreted as an increase in soil acidification.

Regarding N dynamics, no major changes are seen in either deposition or stream water output. Furthermore, as indicated by the data from the catchment Markungsgraben, disturbance of ecosystem balance by, for example, bark beetle infestation will lead to increased N and Al leaching which causes temporarily higher requirement on water treatment plants.

With respect to biological communities, no major recovery is observed yet. However, small changes in population dynamics might be interpreted as the first signs of recovery.

A significant delay in recovery from acidification in the low mountain ranges of Germany has important implications for water authorities, forest managers and policy makers. Forest management is still confronted with the need for frequent liming as well as consideration of sustainable management to avoid base cation depletion of soils and subsequent nutrient deficiencies in the trees. Water authorities should be aware that acidification reversal and recovery of aquatic systems in the area will be delayed, potentially for decades. Even catchment liming results in a very delayed response of stream water in the catchments with deeply weathered soils as demonstrated at the Steile Bramke. Policy makers should consider that N deposition has to be reduced considerably in order to limit cation depletion from soils, risks connected to increasing N saturation and further soil and water acidification.

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