Biogeochemistry **47:** 319–333, 1999.

Patterns of stable S isotopes in a forested catchment as indicators for biological S turnover

CHRISTINE ALEWELL[1] & MATTHIAS GEHRE[2]
[1]*BITÖK, University of Bayreuth, 95440 Bayreuth, Germany;* [2]*UFZ-Center for Environmental Research Leipzig-Halle, 04318 Leipzig, Germany*

Received 6 February 1998; accepted 12 January 1999

Key words: forested catchments, stable S isotopes, SO_4^{2-} reduction, S cycling, S mineralization

Abstract. Despite intensive biogeochemical research during the last thirty years, the relative importance of biological S turnover for the overall SO_4^{2-} budget of forested catchments remains uncertain. The objective of the present study was (i) to gain new insight into the S cycle of the *Lehstenbach* catchment (Northeastern Bavaria, Germany) through the analysis of stable isotopes of S and (ii) to differentiate between sites which are 'hot spots' for SO_4^{2-} reduction and sites where mineralization and adsorption/desorption processes are more important. The $\delta^{34}S$ values and SO_4^{2-} concentrations of soil solutions, throughfall and groundwater at four different sites as well as runoff of the catchment were measured. The relatively low variability of $\delta^{34}S$ in throughfall and bulk precipitation was in contrast to the high temporal and spatial variability of $\delta^{34}S$ in the soil solution. Sulfate in the soil solution of upland sites was slightly depleted in ^{34}S compared to input values. This was most likely due to S mineralization. Sulfate in the soil solution from wetland soils was clearly enriched in ^{34}S, indicating dissimilatory SO_4^{2-} reduction. The observed spatial and temporal patterns of ^{34}S turnover and SO_4^{2-} concentrations might explain the overall balanced S budget of the catchment. At a time of decreasing anthropogenic deposition SO_4^{2-} is currently released from upland soils. Furthermore, mineralization of organic S may contribute to SO_4^{2-} release. Wetland soils in the catchment represent a sink for SO_4^{2-} due to dissimilatory SO_4^{2-} reduction.

Introduction

Biogeochemical research during the last thirty years has improved considerably our understanding of S cycling in forest ecosystems. However, important questions remain unresolved that hinder our ability to understand and predict ecosystem behavior. Most notably, the importance of the biological S turnover in forest ecosystems (mainly the relative contribution of S mineralization and SO_4^{2-} reduction) versus the importance of SO_4^{2-} adsorption/desorption processes are under discussion (e.g. David et al. 1984; Mitchell et al. 1985;

Torssander & Mörth 1997). Some authors suggested that the organic S pool is rather stable and chemical processes dominate S dynamic and fluxes in forest ecosystems (McLaren et al. 1985; Mitchell et al. 1989). Others studies concluded that in ecosystems with ≤ 10 kg S ha^{-1} yr^{-1} deposition nearly all of the deposited SO_4^{2-} cycles through the biomass (David et al. 1984, 1987; Mayer et al. 1995a; Alewell et al. 1999).

The use of stable isotopes in evaluating the S biogeochemistry of forested ecosystems is a promising tool and has been reviewed recently by Mitchell et al. (1998). The isotopic composition in forest ecosystems is controlled by isotopic composition of sources (i.e., atmospheric deposition and mineral weathering) and isotopic discrimination during S transformations. Only biological processes cause shifts in S isotope ratios under natural environmental conditions (Krouse & Grinenko 1991). Generally, bacteria prefer the lighter ^{32}S which causes a depletion of ^{34}S in the reaction product and an enrichment in the remaining solution. Especially dissimilatory SO_4^{2-} reduction results in a major depletion of ^{34}S in the reaction product (Krouse & Grinenko 1991). Recently, other biological processes like S immobilization and mineralization are in discussion to fractionate between the isotopes (Mayer et al. 1995a, b; Mitchell et al. 1998; Alewell et al. 1999). The investigation of the dynamic and patterns of stable S isotopes in forest ecosystem indicates that biological S turnover is an important part of the S cycle in forest soils (Mayer et al. 1995b; Fuller et al. 1986; Alewell et al. 1999).

In the *Lehstenbach* catchment in the Fichtelgebirge (Northeastern Bavaria, Germany) a preliminary study found runoff enriched in $\delta^{34}S$ values compared to the soil solution of an aerobic acid forest soil (Alewell & Giesemann 1996). The conclusion from this study was, that a considerable amount of SO_4^{2-} was reduced in the wetlands of the catchment. However, the average SO_4^{2-} input with deposition between 1993–1996 was 1.03 kmol S ha^{-1} yr^{-1}, and SO_4^{2-} output with runoff was 1.15 kmol S ha^{-1} yr^{-1}. Thus, considering an error margin for flux estimates of 10–20% (Manderscheid & Matzner 1995), the catchment budget is currently balanced or may even indicate net SO_4^{2-} release.

The objectives of this study were (i) to investigate patterns of S stable isotopes in respect to our process understanding at the catchment scale, and (ii) to differentiate between sites which are ‘hot spots’ for SO_4^{2-} reduction and sites where mineralization and adsorption/desorption processes are of greater relevance. The evaluation of differences and similarities between different sites within the catchment was used to improve our general understanding of SO_4^{2-} budgets and S cycling in the *Lehstenbach* catchment.

Site description

The *Lehstenbach* catchment in the *Fichtelgebirge* mountains in north eastern Bavaria (Germany) has an area of 4.2 km^2 with a highest elevation of 877 m a.s.l. Ninety percent of the area is planted by Norway spruce (*Picea abies*, KARST.) of different age classes. Approximately 30% of the catchment is covered by fens or intermittent seeps. Upland soils in the catchment (i.e., oxic soils, which are not water saturated) have developed from weathered granitic bedrock and are predominantly Cambisols and Cambic Podzols (FAO).

The sites *Coulissenhieb* (Figure 1) and *Gemoes* are located in upland areas and support approximately 140 and 110 year old forests, respectively (Figure 1). The site *Weidenbrunnen* is located next to the *Coulissenhieb* but trees are younger in age (about 45 years). *Köhlerloh* is an intermittent seep and trees are approximately 55 years old. This site is often waterlogged in autumn, winter and spring and the ground water is found at less than 1 m depth for the rest of the year.

The annual average precipitation in the catchment between 1995 and 1997 was 1070 mm·yr^{-1}. The average annual temperature was 5 °C.

Methods

Samples were collected monthly from the 4 different sites within the catchment differing in tree age, canopy structure or hydrological conditions. Each site is subdivided into several plots. To determine the spatial variability within sites, throughfall samplers as well as lysimeters were analyzed separately for several sampling events. To determine the variability between sites within the catchment over the two year measurement period, we had to composite samples, because the analysis of δ^{34}S requires a relatively large sample volume and measurements are expensive. If the analysis of variance (one factor *ANOVA*) indicated that the factor *'spatial position'* had no influence on the variability of δ^{34}S values in the solutions and standard error was <0.3‰ (= measurement precision of the mass spectrometer), samples within a site were combined to one composite sample.

Sampling at the *Coulissenhieb* began in August 1995. Soil solution samples as well as throughfall were collected from 4 different measurement plots (20 m^2) within the site. Each plot had 5 suction lysimeters at 90 cm depth and 5 throughfall collectors. The distance between plots was between 100 and 200 m. At a single sampling event each throughfall collector and at 4 sampling events each lysimeter was sampled and analyzed separately to evaluate variability. From then on the 5 samples from each plot were combined volumetrically in one composite sample. Because the analysis of variance

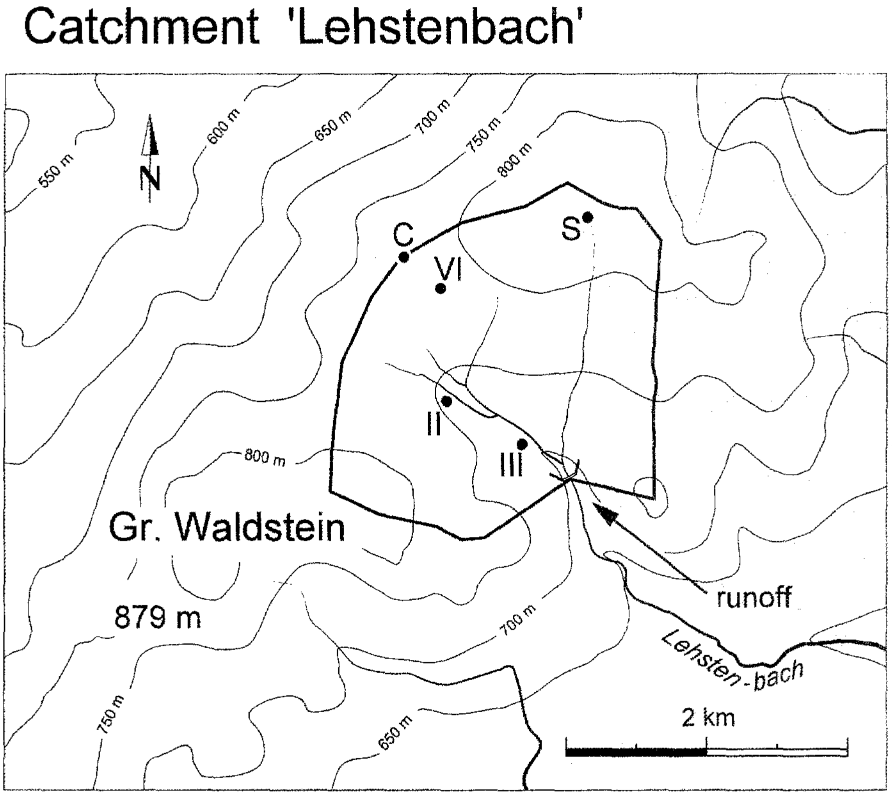

Figure 1. The *Lehstenbach* catchment in the *Fichtelgebirge* mountains. C = *Coulissenhieb*, S = *Bergkopf* spring, VI = *Weidenbrunnen*, II = *Koehlerloh*, III = *Gemoes*.

as well as the standard deviation indicated that spatial variability of $\delta^{34}S$ in throughfall within the site *Coulissenhieb* was low (no influence of the factor position and standard error <0.4‰), all 20 throughfall collectors at the site were combined volumetrically to one composite sample from April 1996 to August 1997.

Bulk deposition was sampled near the site *Coulissenhieb*. Samples from 5 deposition samplers which were installed on a plot of 20 m^2 with a distance of 2 m were combined volumetrically to one composite sample.

Beginning in December 1995, three additional sites (*Köhlerloh*, *Gemoes* and *Weidenbrunnen*) were sampled. At each site, 3 plots with 5 throughfall samplers were implemented. Plots were 10 m apart and within a plot throughfall samplers were installed with a distance of 2 m. For the first 4 sampling events, throughfall samples were combined per plot. Because the analysis of variance as well as the standard deviation indicated a low spatial variability and no influence of position within the sites, all 15 throughfall samplers were combined to one composite sample per site from there on. Each site had one lysimeter plot (6 m^2) with 4 lysimeters at 100 cm depth. The 4 lysimeter samples of each plot were combined to one composite sample. Ground water wells on *Köhlerloh*, *Gemoes* and *Weidenbrunnen* were sampled monthly, starting in November 1995. Ground water wells were sampled depth integrated down to the maximum penetration depth, which was 10, 15 and 16 m for the sites *Köhlerloh*, *Gemoes* and *Weidenbrunnen*, respectively. From January to March 1996 weather conditions did not allow sampling of the wells.

Surface waters in the catchment were sampled monthly beginning September 1995. Because two streams combine directly before the catchment output, they were sampled separately, called runoff 1 and runoff 2, draining the western and the eastern part of the catchment, respectively (Figure 1). Samples of the '*Bergkopf*' spring, which drains mainly upland areas, as well as from a creek draining a fen, were taken in addition.

Sulfate in the solutions was precipitated as $BaSO_4$ by adding $BaCl_2$. The samples were prepared by mixing 1 portion $BaSO_4$ and 5 portions V_2O_5 and encased in tin-foil. The S isotopic composition of the $BaSO_4$ was measured on-line with an elemental analyzer (Carlo Erba 1108) coupled to an isotope ratio mass spectrometer (Finnigan MAT delta S). $\delta^{34}S$ is defined as

$$\delta^{34}S[‰] = [(^{34}S/^{32}S_{\text{sample}})/(^{34}S/^{32}S_{\text{standard}}) - 1]*1000.$$

Troilite from the Canyon Diablo meteorite (CDT) was used as a standard, which has $^{34}S/^{32}S$ ratio of 1/22.22. Precision of the $\delta^{34}S$ measurement was $\pm 0.3‰$.

Statistical analysis of the data was done with *Statistical Package for the Social Sciences (SPSS)*. The Kolmogorov–Smirnov test indicated that data sets came from a normal distribution. To test for significant differences *t*-tests were done. For the analysis of variance one factor *ANOVA* was carried out. All statistical tests were applied over the whole measurement period.

Results and discussion

$\delta^{34}S$ values in precipitation and throughfall

Spatial variability of $\delta^{34}S$ values in throughfall was low. The one factor *ANOVA* indicated that the factor '*spatial position*' had no influence on $\delta^{34}S$ values in throughfall. This was valid for different plots within one site as well as for different sites within the *Lehstenbach* catchment. Standard error of $\delta^{34}S$ values in throughfall within the sites *Coulissenhieb*, *Köhlerloh*, *Gemoes*

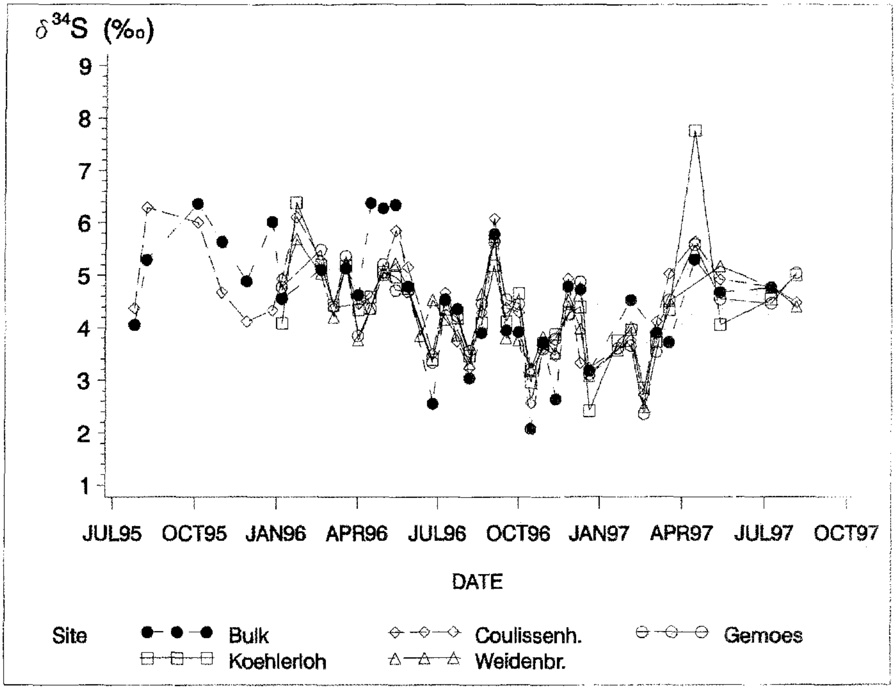

Figure 2. δ^{34}S in throughfall of the sites *Coulissenhieb*, *Koehlerloh*, *Gemoes* and *Weidenbrunnen* as well as bulk precipitation (Bulk) in the Lehstenbach Catchment.

and *Weidenbrunnen* was <0.2‰. Standard error of δ^{34}S in throughfall for the whole catchment was <0.1‰. The latter was valid for the data set where single throughfall samplers were analyzed separately (n = 74; mean = 4.8‰) as well as for the data set where samples were combined within the sites (n = 169; mean = 4.4‰). There was no significant difference between bulk precipitation and throughfall values (Figure 2).

δ^{34}S of neither throughfall nor bulk precipitation showed seasonality (Figure 2). Because there was neither a difference in δ^{34}S values of throughfall between sites of different tree age or canopy structure nor between throughfall and bulk precipitation, we can assume that no isotopic discrimination occurred during S transformations within the canopy of the trees. Furthermore, δ^{34}S values of wet and dry SO_4^{2-} deposition in our catchment were most likely similar. Zhang et al. (1998) found for deciduous forests in North America that δ^{34}S of throughfall did not differ significantly between sites with different tree species. Stam et al. (1992), Van Stempvoort & Wills (1991) as well as Zhang (1994) measured no significant differences in δ^{34}S values of precipitation, throughfall and stem flow for ecosystems in eastern North America. Mayer et al. (1995a) as well as our data showed the same

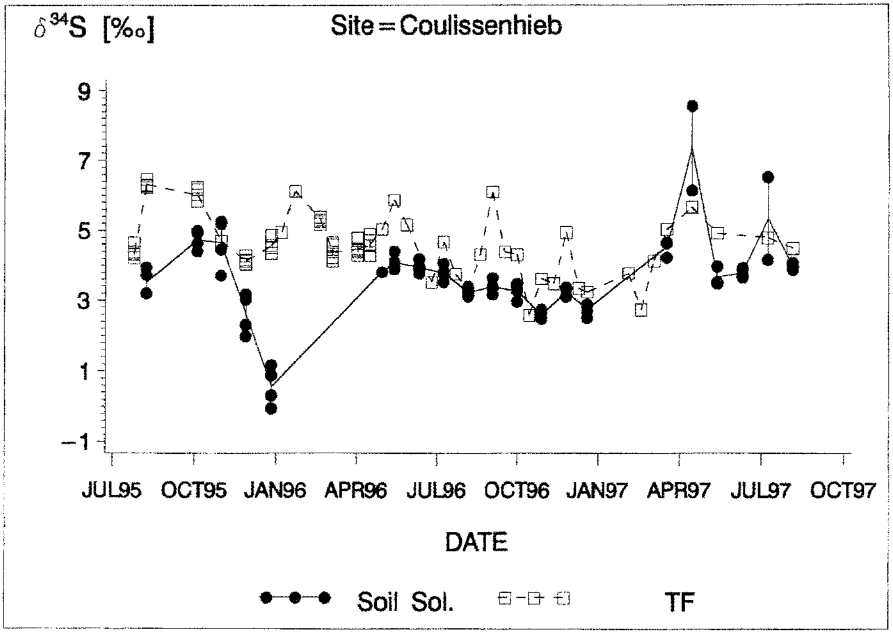

Figure 3. δ^{34}S values in throughfall (TF) and soil solution (Soil Sol.) at the site *Coulissenhieb*. Spatial repetitions are illustrated with equal symbols. Lysimeters were frozen between January and April 1996.

results for spruce sites in Germany during summer and winter, even though dry deposition is an important S source for the catchment.

δ^{34}S values in soil solution indicating biological turnover of S

δ^{34}S values of SO_4^{2-} in soil solutions of the upland sites *Coulissenhieb, Weidenbrunnen* and *Gemoes* were slightly depleted in comparison to SO_4^{2-} in throughfall (Figures 3–5). For the sites *Coulissenhieb* and *Gemoes* this depletion was significant (difference of mean between δ^{34}S in throughfall and soil solution ≥ 1‰, $p < 0.0001$ and $p < 0.001$, respectively). δ^{34}S values of the soil solution at the site *Weidenbrunnen* were only slightly depleted in comparison to throughfall values (difference in mean = 0.4‰) and the difference was not significant.

As mentioned above, the possible SO_4^{2-} sources in the catchment are atmospheric deposition and mineral weathering. Since the bedrock of the catchment is granite, a lithogenic S source is negligible (Mückenhausen 1993). Thus, changes in the isotopic signature from throughfall to soil solutions are due to biological transformations. Several authors in North America and Europe have found that SO_4^{2-} transport in upland forest soils

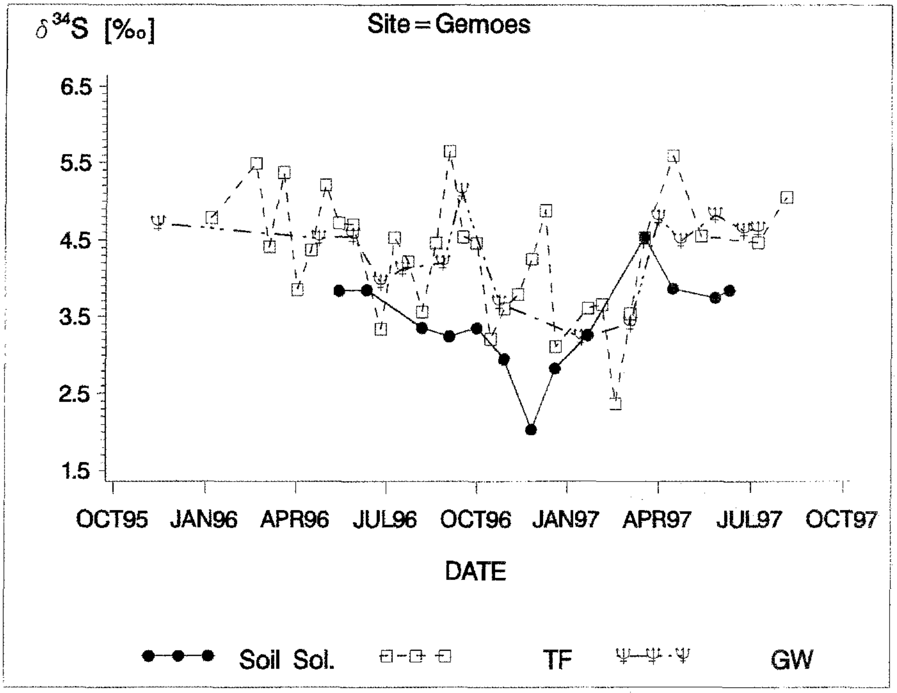

Figure 4. δ^{34}S values in throughfall (TF) and soil solution (Soil Sol.) and ground water (GW) at the site *Gemoes*.

was conservative, because precipitation, soil solution and stream water of forest ecosystems have similar δ^{34}S values (Caron et al. 1986; Stam et al. 1992; Mayer et al. 1995a). However, Mayer et al. (1995a) concluded that even though δ^{34}S values were only slightly altered, the oxygen isotopic composition (δ^{18}O) of SO_4^{2-} indicates that biological processes were contributing to the SO_4^{2-} in soil solution. Recent studies revealed that δ^{34}S values of SO_4^{2-} in soil solutions of upland sites were generally slightly depleted in comparison to SO_4^{2-} in throughfall data (Fuller et al. 1986; Novak et al. 1995; Zhang 1994; Alewell et al. 1999, present data). A depletion in ^{34}S of SO_4^{2-} in soil solution in relation to that in throughfall indicates that S mineralization is a potential SO_4^{2-} source, because the soil microflora prefers the lighter ^{32}S isotope. Furthermore, several authors have shown for North American soils that soil organic S, from which the SO_4^{2-} was released, was depleted in ^{34}S in relation to adsorbed SO_4^{2-} (Fuller et al. 1986; Krouse et al. 1991; Zhang 1994). Lower δ^{34}S values in soil organic matter are likely a result of isotopic discrimination during dissimilatory or assimilatory SO_4^{2-} reduction and incorporation of S in the soil organic matter. However, Mayer et al. (1995a) determined higher δ^{34}S values for the organic S fraction as compared to inorganic soil SO_4^{2-} through-

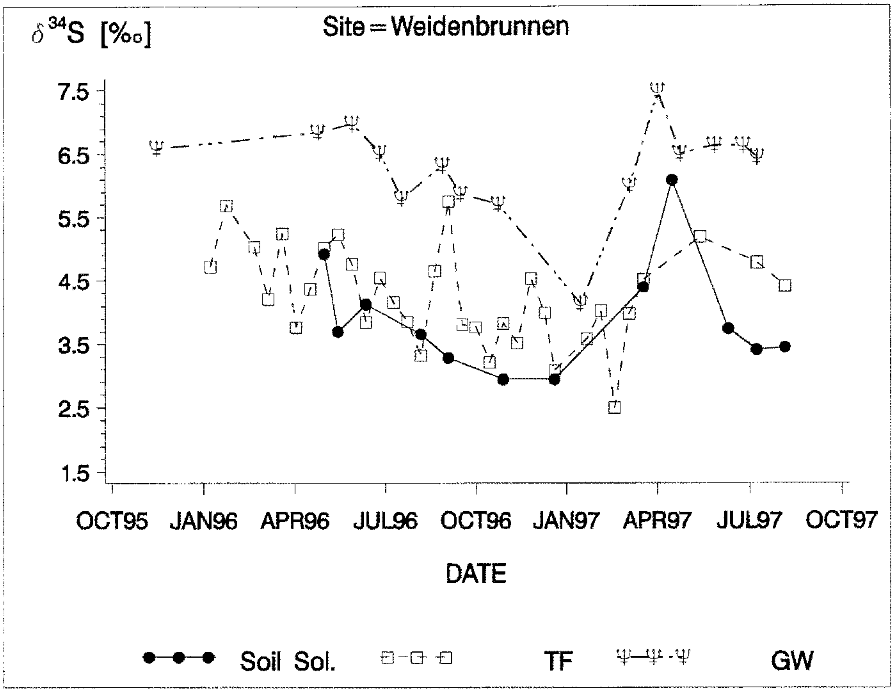

Figure 5. δ^{34}S values in throughfall (TF) and soil solution (Soil Sol.) and ground water (GW) at the site *Weidenbrunnen.*

out the whole soil profile of two soils from the Black Forest in Germany. They concluded that the consistently lower δ^{34}S values in soil solutions were caused by preferential uptake of ^{34}S during immobilization (Mayer et al. 1995a). Overall, authors seemed to agree that the consistently lower δ^{34}S values in soil solutions compared to throughfall and the differences in δ^{34}S between organic and inorganic S at upland forest sites can only be explained by S immobilization/mineralization processes (Fuller et al. 1986; Krouse et al. 1991; Zhang 1994; Mayer et al. 1995a; Alewell et al. 1999).

Sulfate concentrations in soil solutions of the upland sites *Coulissenhieb* and *Gemoes* were distinctly higher than throughfall values (Table 1), which was partly due to evapotranspiration and partly to desorption of inorganic SO_4^{2-} from the soil matrix under decreasing atmospheric SO_4^{2-} deposition (Alewell 1998). Furthermore, the depletion of δ^{34}S in the soil solution points to S mineralization. Whether or not this is a net mineralization of S, cannot be determined with certainty from stable isotope data (see discussion above). The decrease in SO_4^{2-} concentrations in ground water in comparison to soil solution at the upland site *Gemoes* (Table 1) was not caused by SO_4^{2-} reduction, because ground water was not enriched in ^{34}S. A recent study in the

Table 1. Average SO_4^{2-} concentrations (1993–1996) in throughfall (TF), soil solution (Soil Sol.) and ground water (GW) at the investigated sites in $\mu mol \cdot l^{-1}$.

Site	Throughfall	Soil solution	Ground water
Coulissenhieb	126.6	278.3	–
Köhlerloh	166.9	39.5	36.2
Gemoes	150.1	418.6	215.0
Weidenbrunnen	126.8	234.9	89.5

Lehstenbach catchment found that the deeply weathered granite has a considerable SO_4^{2-} adsorption capacity to a depth of over 10 m (Manderscheid et al. 1998). This might be responsible for the lower SO_4^{2-} concentrations of the ground water at the site *Gemoes*.

In contrast to the upland sites, $\delta^{34}S$ values in soil solution at the site *Köhlerloh* (Figure 6) were significantly enriched compared to throughfall values (difference in mean = 4.2‰, $p < 0.001$). Increased $\delta^{34}S$ values in the soil solution suggest dissimilatory SO_4^{2-} reduction in the soil. *Köhlerloh* is an intermittent seep which is often waterlogged in autumn, winter and spring and ground water is found at less than 1 m depth for the rest of the year. Thus, the site has favorable conditions for SO_4^{2-} reduction. Soil solutions of *Köhlerloh* had low SO_4^{2-} concentrations compared to throughfall values, supporting the conclusion that a considerable part of the deposited SO_4^{2-} was reduced at this site. The ground water had intermediate $\delta^{34}S$ values between throughfall and soil solution indicating a temporal and spatial mixture of drainage from reducing and nonreducing areas.

$\delta^{34}S$ values of throughfall and soil solution at *Weidenbrunnen* (Figure 5) were not significantly different but the ground water was significantly enriched in ^{34}S (mean difference between $\delta^{34}S$ in throughfall and ground water = 2‰, $p \leq 0.0001$). This points to dissimilatory SO_4^{2-} reduction zones in depths greater than 1 m. Sulfate concentrations, which were markedly lower in the ground water than in the soil solution, supported this conclusion (Table 1). Soil characteristics at the site *Weidenbrunnen* gave further evidence that SO_4^{2-} reduction was likely to occur in deeper soil layers because there is a gleyic horizon at ≥ 1 m depth.

Even though the conclusions of Alewell and Giesemann (1996) that dissimilatory SO_4^{2-} reduction is an ongoing process in the catchment can be confirmed by this study, it was clear that the process has a relatively high temporal variability. While soil solution data indicated that there was no SO_4^{2-} reduction occurring at *Köhlerloh* in 1997, soil solutions from the upland

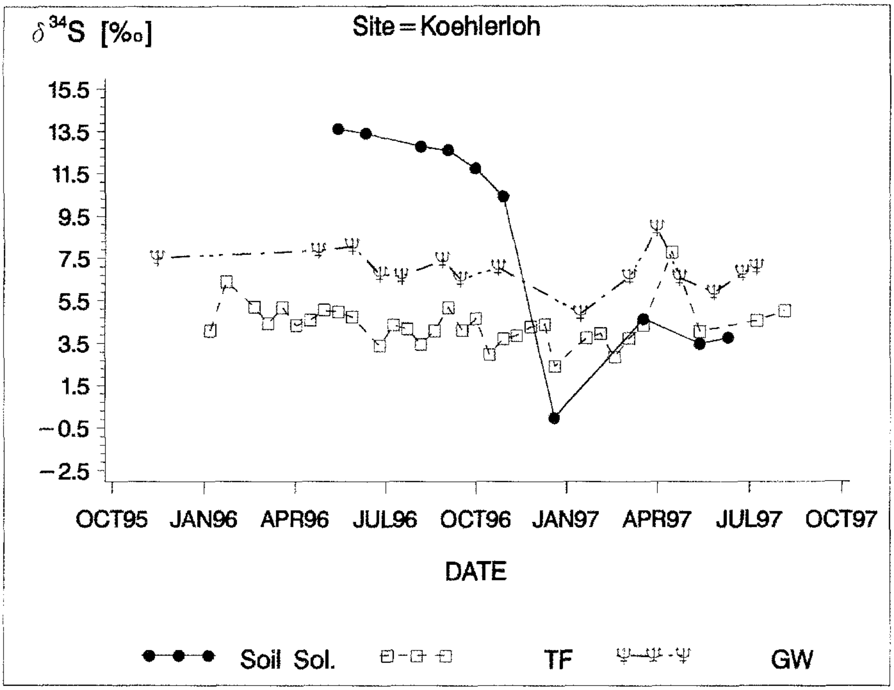

Figure 6. δ^{34}S values in throughfall (TF) and soil solution (Soil Sol.) and ground water (GW) at the site *Koehlerloh*.

site *Coulissenhieb* as well as surface waters, were enriched in ^{34}S during April 1997 (Figures 3–5). Thus, SO_4^{2-} reduction was not totally restricted to wetland areas but may also occur on upland sites.

Temporal variability of δ^{34}S in soil solution was generally the highest of all investigated compartments. However, we did not see a seasonality of δ^{34}S values with two exceptions. In the winter of 1996, δ^{34}S values in soil solution of *Coulissenhieb* as well as surface waters declined about 4‰ (Figures 3 and 5). Soil solutions of *Coulissenhieb* and *Weidenbrunnen* and surface waters of the catchment, showed no change in the winter of 1997. δ^{34}S values in the soil solution of the site *Köhlerloh* declined from 11.5‰ in October 1996 to −0.3‰ in December 1996 and δ^{34}S values in soil solution of *Gemoes* decreased about 1.5‰ (Figures 4 to 6). So far we cannot explain the inconsistent sudden drop in δ^{34}S values in the winter. We might assume an interruption of SO_4^{2-} reduction processes in the winter months and sudden lateral flow of water with different δ^{34}S signature at the intermittent seep *Köhlerloh*. However, these processes could not explain the decline in δ^{34}S values in soil solution at the sites *Coulissenhieb* and *Gemoes*.

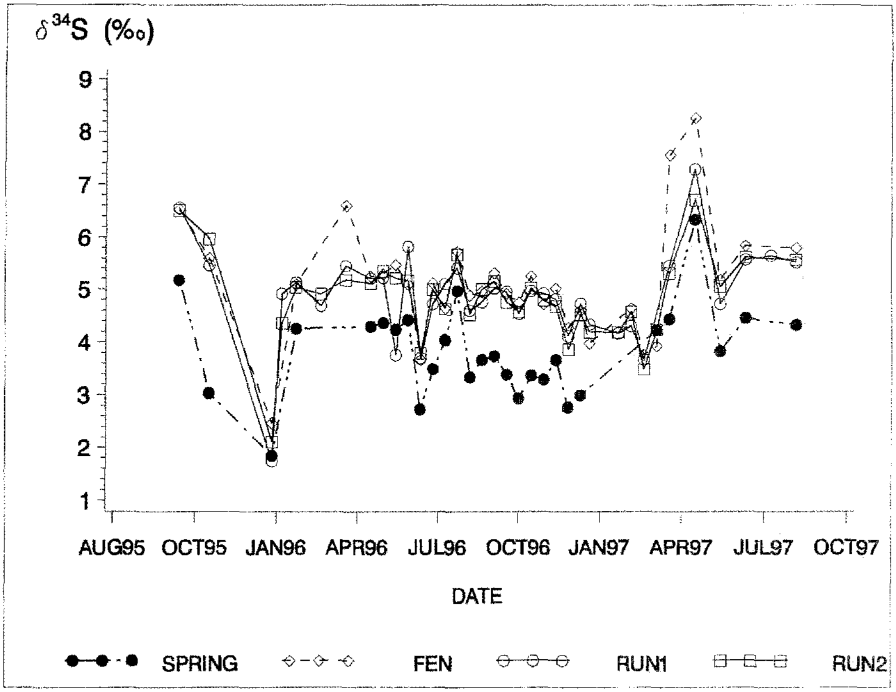

Figure 7. δ^{34}S values in surface waters. SPRING = *Bergkopf* spring, Run1 and Run2 = runoff draining western and eastern parts of the catchment, respectively. FEN = creek draining a fen.

δ^{34}S values in surface waters

δ^{34}S values in surface waters of the catchment showed a low spatial variability (Figure 7). Runoff 1 and runoff 2, which drain the western and the eastern part of the catchment, respectively, were not significantly different in δ^{34}S values (mean values 4.8 and 4.9‰, respectively). δ^{34}S values of the creek draining the fen were only slightly higher (mean δ^{34}S = 5.1‰).

There was a significant difference in δ^{34}S values between soil solution and runoff, with the runoff being significantly higher than the soil solution from the upland sites *Coulissenhieb*, *Gemoes* and *Weidenbrunnen* (difference in mean δ^{34}S = 1.3‰, $p < 0.0001$). Furthermore, δ^{34}S in runoff was significantly higher than throughfall values, even though the difference was small (difference in mean δ^{34}S = 0.5‰, $p < 0.01$). For most periods surface waters were enriched by about 1–2‰ compared to soil solutions of upland sites. This enrichment in ^{34}S of the surface waters indicates ongoing dissimilatory SO_4^{2-} reduction in the wet areas of the catchment. δ^{34}S values of the spring, which drains mainly upland forest soils, were significantly lower than runoff and throughfall values (difference in mean δ^{34}S = 1.2‰ and 0.6‰, $p < 0.0001$

and $p < 0.03$, respectively). As discussed above, a depletion of ^{34}S in drainage waters points to immobilization and mineralization processes.

Conclusions

Since we found low temporal and spatial variability in the δ^{34}S values of the SO_4^{2-} input into the catchment but a large spatial and temporal variability for δ^{34}S values of the soil solution, we conclude that biological turnover in the soil is an important part in the S cycle of our forest ecosystems. Thus, the soil is a reactive pool for SO_4^{2-} which creates variability in stable isotope patterns.

The sites within the *Lehstenbach* catchment had different patterns in their stable isotope data pointing to differences in S metabolism. *Köhlerloh* seemed to be a "hot spot" for SO_4^{2-} reduction. *Coulissenhieb* and *Gemoes* were more influenced by SO_4^{2-} desorption and/or S mineralization. δ^{34}S values of ground water at *Weidenbrunnen* indicated, that SO_4^{2-} reduction in deeper soil layers ($\geq$1 m depth) occurred. The different δ^{34}S patterns at the sites can also explain the balanced SO_4^{2-} budget of the catchment. While SO_4^{2-} was released from upland sites due to S mineralization and desorption of inorganic SO_4^{2-} under decreasing atmospheric inputs, the wet sites of the catchment retained SO_4^{2-} due to reduction processes.

Acknowledgement

This work was supported by the German Federal Ministry of Education, Science, Research and Technology (BMBF), Grant No. BEO 51-0339476B. We would like to thank Jan Irmler for his help with the experimental work and Pat McHale for the language editing of the manuscript.

References

Alewell C & Giesemann A (1996) Sulfate Reduction in a forested catchment as indicated by δ^{34}S of soil solutions and runoff. Isotopes in Environmental and Health Studies 32: 203–210

Alewell C (1998) Investigating sulfate sorption and desorption of acid forest soils with special consideration of soil structure. J. Soil Sci. Plant Nutrition 161: 73–80

Alewell C, Mitchell M, Likens GE & Krouse HR (1999) Sources of stream sulfate at the Hubbard Brook Experimental forest: Long-term analyses using stable isotopes. Biogeochemistry 44: 281–299

Caron F, Tessier A, Kramer JR, Schwarcz HP & Rees CE (1986) Sulfur and oxygen isotopes of sulfur in precipitation and lake water, Quebec, Canada. Appl. Geochem. 1: 601–606

David MB, Mitchell, MJ & Schindler SC (1984) Dynamics of organic and inorganic sulfur constituents in hardwood forest soils. In: Stone EL (Ed) Forest Soils and Treatment Impacts; Sixth North American Forest Soil Conference (pp 221–245). Knoxville, Tenn., U.S.A.

David MB, Mitchell MJ & Scott MT (1987) Importance of biological processes in the sulfur budget of a northern hardwood ecosystem. Biol. Fertil. Soils 5: 258–264

Fuller RD, Mitchell MJ, Krouse HR, Wyskowski BJ & Driscoll CT (1986) Stable sulfur isotope ratios as a tool for interpreting ecosystems sulfur dynamics. Water Air and Soil Pollution 28: 163–171

Krouse HR & Grinenko VA (1991) Stable Isotopes. Natural and Anthropogenic Sulphur in the Environment. Scope 43, John Wiley & Sons Ltd.

Krouse HR, Stewart JWB & Grinenko VA (1991) Pedosphere and Biosphere. In: Krouse HR & Grinenko VA (ED) Stable Isotopes. Natural and Anthropogenic Sulphur in the Environment (pp 267–306). Scope 43, John Wiley & Sons Ltd.

Manderscheid B & Matzner E (1995) Spatial and temporal variability of soil solution chemistry and seepage water ion fluxes in a mature Norway spruce (*Picea abies* (L.) Karst.) stand. Biogeochem. 30: 99–114

Manderscheid B, Schweisser T, Lischeid G, Alewell C & Matzner E (1998) Sulfate pools in the weathered bedrock of a forested catchment. Soil Sci. Soc. Am. J. (submitted)

Mayer B, Feger KH, Giesemann A & Jäger H-J (1995a) Interpretation of sulfur cycling in two catchments in the Black Forest (Germany) using stable sulfur and oxygen isotope data. Biogeochem. 30: 31–58

Mayer B, Fritz P, Prietzel J & Krouse HR (1995b) The use of stable and oxygen isotope ratios for interpreting the mobility of sulfate in aerobic forest soils. Appl. Geochem. 10: 161–173

McLaren RG, Keer JI & Swift RS (1985) Sulphur transformations in soils using sulphur-35 labeling. Soil Biol. Biochem. 17: 73–79

Mitchell MJ, Driscoll CT, Fuller RD, David MB & Likens GE (1989) Effect of whole-tree harvesting on the sulfur dynamics of a forest soil. Soil Sci. Soc. Am. J. 53: 933–940

Mitchell MJ, David MB, Maynard DG & Telang SA (1985) Sulfur constituents in soils and streams of a watershed in the Rocky Mountains of Alberta. Can. J. For. Res. 16: 315–320

Mitchell MJ, Krouse HR, Mayer B, Stam AC & Zhang YM (1998) Use of stable isotopes in evaluating biogeochemistry of forest ecosystems. In: McDonnell & Kendall C (Ed) Isotope Tracers in Catchment Hydrology. Elsevier, The Netherlands (in press)

Mückenhausen E (1993) Die Bodenkunde und ihre geologischen, geomorphologischen, mineralogischen und petrologischen Grundlagen. DLG-Verlag Frankfurt am Main, 1-579

Novak M, Bottrell SH, Groscheova H, Buzek F & Cerny J (1995) Sulphur isotope characteristics of two North Bohemian forest catchments. Water Air and Soil Pollution 85: 1641–1646

Stam AC, Mitchell MJ, Krouse HR & Kahl JS (1992) Stable sulfur isotopes of sulfate in precipitation and stream solutions in a northern hardwood watershed. Water Resour. Res. 28: 231–236

Van Stempvoort DR & Wills JJ (1991) Aboveground vegetation effects on the deposition and cycling of atmospheric sulfur: Chemical and stable isotope evidence. Water Air and Soil Pollution 60: 55–82

Torssander P & Mörth C-M (1997) Sulfur dynamics in the roof experiment at Lake Gårdsjön deduced from sulfur and oxygen isotope ratios in sulfate. In: Hultberg H & Skeffington R (Eds) Experimental Reversal of Acid Rain Effects: The Gårdsjön Roof Project (pp 1–466). John Wiley & Sons, Chichester, U.K.

Zhang Y (1994) Biogeochemistry of forest soils at the Hubbard Brook Experimental Forest, New Hampshire: an experimental approach. PhD Dissertation. SUNY, College of Environmental Science and Forestry, Syracuse, NY, U.S.A.
Zhang Y, Mitchell MJ, Christ M, Likens GE & Krouse HR (1998): Stable sulfur isotopes at the Hubbard Brook Experimental Forest, New Hampshire. Biogeochem. (in press)