

USE OF NATURAL ^{35}S TO TRACE SULPHATE CYCLING IN SMALL LAKES, FLATTOPS WILDERNESS AREA, COLORADO, U.S.A.

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Abstract. Measurements of the cosmogenically-produced ^{35}S , a radioisotope of sulphur ($t_{1/2} = 87$ days), are reported for the Ned Wilson Lake watershed in Colorado. The watershed contains two small lakes and a flowing spring presumed to be representative of local ground water. The watershed is located in the Flattops Wilderness Area and the waters in the system have low alkalinity, making them sensitive to increases in acid and sulphate deposition. Time series of ^{35}S measurements were made during the summers of 1995 and 1996 (July–September) at all three sites. The system is dominated by melting snow and an initial concentration of $16\text{--}20 \text{ mBq L}^{-1}$ was estimated for snow-melt based on a series of snow samples collected in the Rocky Mountains. The two lakes had large initial ^{35}S concentrations in July, indicating that a large fraction of the lake water and sulphate was introduced by meltwater from that year's snowpack. In 1995 and 1996, ^{35}S concentrations decreased more rapidly than could be accounted for by decay, indicating that other processes were affecting ^{35}S concentrations. The most likely explanation is that exchange with sediments or the biota was removing ^{35}S from the lake and replacing it with older sulphate devoid of ^{35}S . In September of 1995 and 1996, ^{35}S concentrations increased, suggesting that atmospheric deposition is important in the sulphate flux of these lakes in late summer. Sulphur-35 concentrations in the spring water were highly variable but never higher than 3.6 mBq L^{-1} and averaged 2 mBq L^{-1} . Using a simple mixing model, it was estimated that 75% of the spring water was derived from precipitation of previous years.

Keywords: acid deposition, hydrology, mountain lakes, radioactivity, sulphur

1. Introduction

Acidification caused by atmospheric deposition has received extensive attention during the past three decades because of the potential for damage to ecosystems (Delleur, 1989; Weider *et al.*, 1998). Specifically, sulphate and nitrate, produced during combustion of fossil fuels, have produced serious ecological damage over large areas of the industrialized world. The production and release of SO_2 to the atmosphere by combustion of coal and oil in power plants has been destructive to watersheds in large areas of Europe and the eastern United States. The SO_2 can either be deposited as dry deposition or be oxidized to sulphate and deposited in precipitation. Some of the damage caused by sulphate deposition is local, as power plants are point sources of pollution and most of the sulphate may deposit near



the point of origin. However, in the northeastern United States and Appalachian region, most of the damage has resulted from the influx of sulphate from power-producing plants in the mid-western United States. These power plants, which have high emission stacks for dispersal of flue gasses, can pollute areas 200–300 miles downwind of their location. The resulting acidification has caused damage to ecosystems and loss of species diversity, particularly for fish and amphibians (Weider *et al.*, 1998). Emission restrictions have resulted in a decrease in the release of SO₂ and resulted in an improvement in the acidification problem for some hydrologic systems. However, in systems with very low alkalinity and a limited capacity to recover from acidification, it is unlikely that recovery will be possible, even if emissions were decreased to well below predicted levels (Webb *et al.*, 2000).

One method for tracing the effects and movement of atmospheric sulphur in watersheds is through the use of natural sulphur-35. Sulphur-35 is a radioactive isotope of sulphur with a half-life of about 87 days. It is produced naturally in the atmosphere by the spallation of argon atoms by cosmic-rays (Lal and Peters, 1966). It rapidly oxidizes to sulphate and deposits on the Earth's surface either through precipitation or dry fallout (Tanaka and Turekian, 1995). Once on the Earth's surface, it will follow the pathway of atmospherically deposited sulphur through the hydrologic cycle. In hydrologic systems where sulphur is conservative, it can be used as a dating tool for young water (Sueker *et al.*, 1999). In all cases, it serves as a tracer for the paths and timescales of movement of atmospheric sulphur through the system (Michel *et al.*, 2000).

2. Site Description and Chemistry

One area in the United States that may be susceptible to irreversible acidification is the Flattops Wilderness Area, located in northwestern Colorado. The Flattops watersheds have been declared a class 1 wilderness area which means they are to be protected from atmospheric pollution. However, the area is located south of power plants along the Yampa River whose emissions could increase acidity in the Rocky Mountains. The area is also located east of the Piceance Basin which contains large deposits of oil shale. If development of these resources becomes economically feasible in the future, sulphate generation could have a negative impact on the area despite the legal safeguards.

The Flattops Wilderness Area consists of a cap formed by the weathering of a uniform Tertiary basalt layer which forms a mesa at an altitude of about 3350 m. The basalt cap is heavily fractured and has a thin soil layer with vegetation consisting of sub-alpine grasses, alpine willow, and forests of spruce and fir. Due to the fracturing of the basalt, there is very little surface runoff within the area, but a large number of depressions exist which form a series of small lakes and ponds fed by inflow from the small surrounding basins. Ground water is thought to consist mostly of recent precipitation which flows through the overlying soil system into

the fractured basalt. Due to the limited soil development and the nature of the bedrock, most of the lakes in the Flattops have low alkalinity and are susceptible to degradation by atmospheric deposition (Turk and Adams, 1983).

The U.S. Geological Survey has been studying this area for the past two decades. The Ned Wilson Lake watershed (Figure 1) was chosen as a representative site for long-term monitoring following early synoptic studies (Turk and Adams, 1983). The watershed consists of three sampling sites: Ned Wilson Lake (NWL), a pond above Ned Wilson Lake referred to as Upper Ned Wilson Lake (UNWL) here, and a small spring located below NWL that flows throughout the summer. Precipitation at this site averages $1\text{--}1.5\text{ m yr}^{-1}$, as determined by snow pack measurements during spring and weighing-bucket collectors for the rest of the year (Campbell *et al.*, 1991). Summer evaporation is estimated to be less than one quarter of this amount (Spahr and Turk, 1985). The NWL watershed has a surface drainage of approximately 50 ha and is located at an average altitude of 3389 m. Ned Wilson Lake has a surface area of 1 ha and an average depth of 5.5 m. Because most precipitation recharges ground water instead of becoming surface water, flow of surface water into or out of the lake is minimal with regard to the water balance. Based on lake volume and precipitation, the residence time for water in NWL was estimated to be 2–4 yr (Campbell *et al.*, 1991). This residence time estimate does not take into account any flow of ground water into or out of the lake. UNWL, located on a shelf just above NWL, has a surface area of 0.26 ha and a watershed of about 0.5 ha. The average depth of the pond is 1.5 m, similar to the direct precipitation for the area. Due to its location, inflow from ground water should not be a factor at UNWL, although there may be some outflow to ground water. The water in UNWL is thought to reflect yearly precipitation. The spring is located below NWL and flows throughout the summer. It is thought to be representative of the shallow ground water in the area. Due to the lack of a significant drainage, the ground water is presumed to be very young and tritium measurements made in 1992 showed concentrations of about 10–11 T.U. at all three sampling locations, similar to tritium concentrations in precipitation during that period (Michel and Turk, 1996).

The chemistry of the watershed has been monitored by the U.S. Geological Survey since 1982. The chemistry in the system is controlled by a combination of weathering, precipitation chemistry, and evaporation. Turk and Spahr (1991) have found that dry deposition is not a major factor in the chemistry of the watershed. Weathering appears to be the primary control for cations like calcium and magnesium, and for bicarbonate. As a result, their concentrations in the spring water are several times the concentrations in precipitation. The alkalinities are extremely low in this watershed ($<70\ \mu\text{eq L}^{-1}$) making it particularly vulnerable if acid deposition increases in the future. For chloride and sulphate, the primary control seems to be atmospheric deposition, with concentrations in spring water being slightly greater than the concentrations in lake water. The controls on chemical concentrations in precipitation in the Rocky Mountains are complex and not easily understood. There

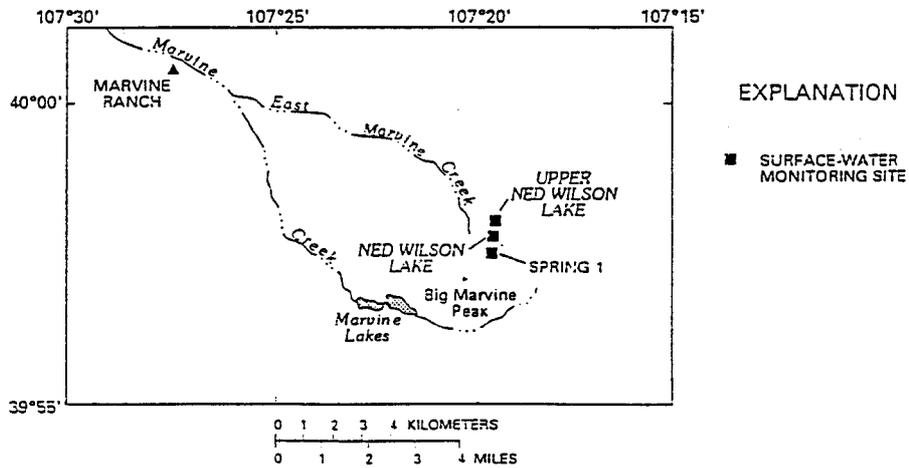
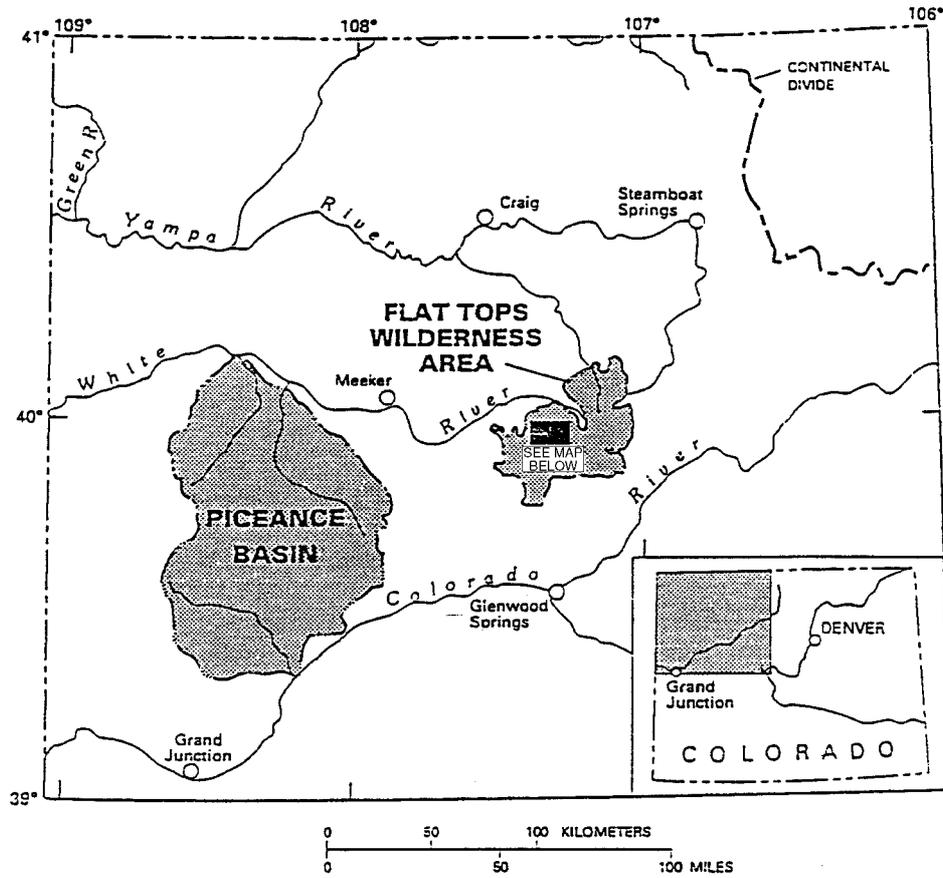


Figure 1. Location of Flattops Wilderness Area and sampling sites in Ned Wilson Lake Watershed.

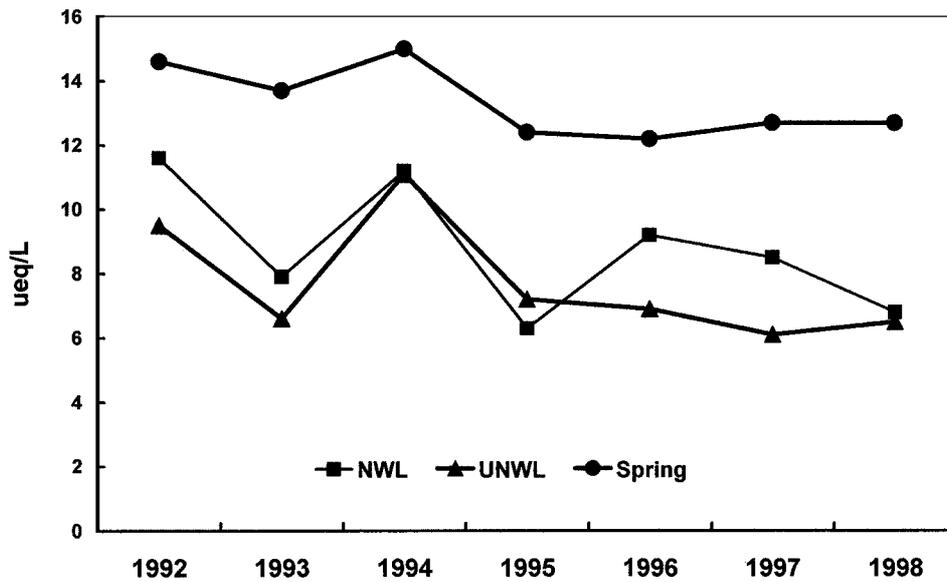


Figure 2. Average sulphate concentration in $\mu\text{eq L}^{-1}$ for the three sites at the Ned Wilson Lake Watershed, 1992–1998.

is evidence from stable sulphur isotopic data that some sulphate being deposited in the Rockies is the result of power-plant generation (Turk *et al.*, 1992). Campbell *et al.* (1991) found that sulphate concentrations in UNWL followed the same trends as concentrations in precipitation. Sulphate concentrations in NWL were related to precipitation plus residual lake water from the previous year. The concentrations of sulphate in the spring water appeared to be controlled by more complex processes, or by processes that occur on longer timescales. The average sulphate concentrations measured at the three sites during 1992–1998 are given in Figure 2. Sulphate concentration in the spring water is about a factor of two higher than in the two lakes.

3. Sample Collection and Analysis

Samples for ^{35}S measurements and water chemistry were collected from all three sites (when accessible) during the summers of 1995–1996 and from the lakes in 1997. Samples were obtained from a well-mixed site along the shore of the lakes. Twenty liters of water were passed through anion exchange columns which were then returned to the laboratory for processing. Due to the low sulphate concentrations, approximately 0.1 g of sodium sulphate known to be ^{35}S free was added to each sample as a carrier. The samples were eluted with 3 M NaCl, and the sulphate was precipitated by the addition of an excess of barium chloride. The precipitate

TABLE I

Sulphur-35 data for sites at Ned Wilson Lake watershed, 1995–1997. ^{35}S data given in both mBq L^{-1} and normalized to sulphate ($\text{mBq mg}^{-1}\text{SO}_4$). Julian dates given in parenthesis.

Date	$^{35}\text{S}\pm 1\sigma$ (mBq L^{-1})	SO_4 ($\mu\text{eq L}^{-1}$)	$^{35}\text{S}/\text{SO}_4$ (mBq mg^{-1})	Inventory ($\text{Bq} \times 10^4$)	^{34}S (‰)
<i>Ned Wilson Lake</i>					
7/26/1995 (207)	8.2±0.6	7.7	22.2	45	5.2
8/15/1995 (227)	2.9±0.5	5.6	10.8	16	
8/30/1995 (242)	0.8±0.4	4.4	3.8	4.4	
9/19/1995 (262)	8.0±1.8	5.8	28.7	44	
9/27/1995 (270)	7.6±1.7	7.1	22.3	42	
7/11/1996 (193)	8.1±0.6	10.8	15.6	45	5.3
8/08/1996 (221)	6.7±1.1	8.3	16.8	37	
8/29/1996 (242)	4.1±1.3	9.0	9.5	23	4.3
9/20/1996 (259)	6.3±1.4	9.0	14.6	35	4.5
7/23/1997 (204)	6.0±0.7	8.5	14.7	33	
8/28/1997 (240)	5.6±0.6	8.7	12.2	31	
9/16/1997 (259)	4.8±0.5	7.6	13.2	21	
<i>Upper Ned Wilson Lake</i>					
7/26/1995 (207)	11.4±0.6	6.5	36.5	4.3	4.7
8/15/1995 (227)	5.2±0.5	6.7	16.2	2.0	
8/30/1995 (242)	4.1±0.4	7.5	11.4	1.5	
9/19/1995 (262)	8.3±1.8	10.0	17.3	3.1	
9/27/1995 (270)	5.0±1.9	10.6	9.8	1.9	
7/11/1996 (193)	8.0±0.6	5.3	31.4	3.0	
8/08/1996 (221)	6.5±1.2	5.6	24.2	2.4	4.8
8/29/1996 (242)	2.7±1.5	6.3	8.9	1.0	
9/20/1996 (259)	5.3±1.5	7.1	15.6	2.0	
8/27/1997 (239)	5.2±0.6	6.2	17.5	2.0	
<i>Spring</i>					
7/26/1995 (206)	2.6±0.6	11.9	4.6		5.1
8/15/1995 (227)	1.5±0.5	12.3	2.5		
8/30/1995 (242)	1.0±0.4	14.0	1.5		
9/19/1995 (262)	3.3±1.8	12.3	5.6		
9/27/1995 (270)	1.9±1.8	13.3	3.0		
7/11/1996 (193)	2.9±0.8	10.8	5.6		
8/08/1996 (221)	3.6±1.0	12.3	6.1		
8/29/1996 (242)	1.3±1.3	12.6	2.1		
9/20/1996 (264)	0.5±1.3	12.4	0.8		

TABLE II

Impact of biogeochemical processes on sulphate chemistry in Ned Wilson Lake Watershed. Processes are decay, mineral weathering, exchange with sediment or biota, biological uptake, groundwater inflow, atmospheric deposition, microbial reduction and evaporation. Processes can either increase (I), lower (L) or have no impact (0) on a given parameter. Note that the impact on ^{34}S will either make it more positive (I) or less positive (L) or cause no change (0)

Processes	^{35}S	SO_4	$^{35}\text{S}/\text{SO}_4$	^{34}S
Decay	L	0	L	0
Weathering of minerals	0	I	L	L
Biogeochemic exchange	L	0	L	0
Biological uptake	L	L	0	0
Groundwater inflow	0	I	L	0
Atmospheric deposition	I	0	I	0
Reduction	L	L	L	L
Evaporation	I	I	0	0

was then analyzed as described in Michel *et al.* (2000) at the U.S. Geological Survey Tritium Laboratory. Results of the analyses, corrected to the date of collection are given in Table I. All results are given both in mBq L^{-1} with 1σ uncertainties and normalized to sulphate ($\text{mBq mg}^{-1}\text{SO}_4$). Inventories of ^{35}S in Becquerels for the lakes are also tabulated. For some samples, ^{34}S of sulphate was measured and these results are given in ‰ in Table I (Mast *et al.*, 2001).

4. Sulphur Isotope Chemistry in the Ned Wilson Lake Watershed

Various processes impact the concentrations of sulphate, ^{35}S , the $^{35}\text{S mg}^{-1}\text{SO}_4$ ratio, and the stable sulphur isotopic ratio in lake water. The possible processes in this watershed are: (1) radioactive decay; (2) mineral weathering within the watershed; (3) exchange with the sediments or biota; (4) biological uptake; (5) microbial reduction of sulphate; (6) atmospheric deposition; (7) inflow of ground water; and (8) evaporation. The impacts of each process on the concentrations and isotopic ratios of sulphate in the NWL watershed are given in Table II.

Several processes (decay, biogeochemical exchange, biological uptake or reduction, and ground water inflow) decrease the ^{35}S concentration in lake water. However, these processes have different impacts on the other properties of sulphur. Decay and biogeochemical exchange decrease both ^{35}S concentration and the $^{35}\text{S mg}^{-1}\text{SO}_4$ ratio but have no effect on sulphate concentrations or the stable

isotopic ratios. Biological uptake and reduction change sulphate concentrations in the same manner as ^{35}S concentrations, leaving the $^{35}\text{S mg}^{-1}\text{SO}_4$ ratio unchanged. Biological reduction of sulphate, unlike uptake, will also have an impact on ^{34}S isotopic ratios. Ground water inflow increases sulphate concentration but not ^{35}S concentrations, therefore decreasing the $^{35}\text{S mg}^{-1}\text{SO}_4$ ratio. It will also result in an increase in cations. Only evaporation and atmospheric deposition increase ^{35}S concentrations. In this watershed, atmospheric deposition will increase the ^{35}S concentrations but not sulphate concentrations, thus increasing the $^{35}\text{S mg}^{-1}\text{SO}_4$ ratio. Evaporation will increase both sulphate and ^{35}S , leaving the $^{35}\text{S mg}^{-1}\text{SO}_4$ ratio the same.

In the Ned Wilson Lake watershed, stable isotope ratios show little change as sulphate moves through the environment. Measurements of ^{34}S in snow are about +4.5–5.5‰ (Mast *et al.*, 2001), which is very similar to the isotopic values found in the surface waters in the watershed (Table I). Thus, processes impacting the stable isotopic ratios, such as reduction of sulphate to sulphide or weathering of sulphide, are not important in this system because of the lack of significant shifts in the ^{34}S ratios (Krouse and Mayer, 2000). Processes such as ground water input or evaporation are not important in the lakes because there is no increase in concentrations of sulphate and other dissolved constituents. The lack of a notable decrease in sulphate concentration indicates that biological uptake is not a major component of the sulphate flux. Thus, the main processes that appear to change ^{35}S concentrations in these lakes are decay, biogeochemical exchange, and addition of ^{35}S by atmospheric deposition. Measurements of ^{35}S in the lake water will give information on the relative importance and rates of these hydrologic processes.

5. Sulphur-35 in Snowpack and Precipitation

Concentrations of ^{35}S in precipitation are highly variable, due to a variety of atmospheric phenomena (Tanaka and Turekian, 1995). Figure 3 shows the ^{35}S concentrations measured in snowpack and precipitation from a variety of locations within the Rocky Mountains (Michel *et al.*, 2000). The snowpack samples were full depth composites collected in late spring just prior to snowmelt. The precipitation samples were collected at Loch Vale, Colorado and Deep Creek, Colorado, which are northeast and southeast of the watershed respectively. The ^{35}S concentrations in rainfall can be extremely variable, especially in the late spring and summer. Higher concentrations of ^{35}S , derived from the stratosphere, may be introduced into the troposphere either by the breakup of the tropopause in spring or the penetration of thunderheads through the tropopause.

As the major hydrologic event for the Ned Wilson Lake watershed is snowmelt, the major input of ^{35}S into the system occurs in the spring and early summer when the snowpack melts. As Figure 3 shows, despite being a collector of dryfall as well as wetfall, the snowpack generally has a lower ^{35}S concentration than rain. This

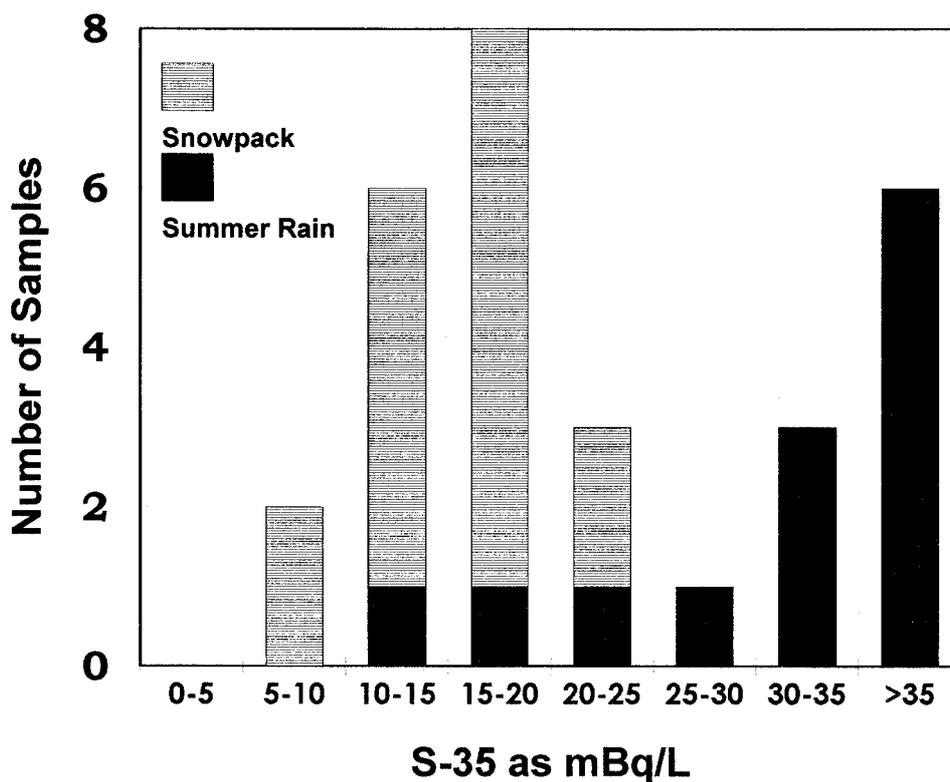


Figure 3. ^{35}S concentrations in mBq L^{-1} in snowpack and summer rain from sites in the Rocky Mountains (Michel *et al.*, 2000).

is primarily due to the fact that the snowfall accumulates over a period of months, so much of the ^{35}S that falls in the autumn and early winter will have decayed before spring melt. The average ^{35}S concentration for snowpack collected in the Rocky Mountains is $16 \pm 4 \text{ mBq L}^{-1}$ (Michel *et al.*, 2000). The two snowpack samples collected at the Ned Wilson Lake watershed had concentrations of 14.0 (March 31, 1994) and 20.2 mBq L^{-1} (June 2, 1995). Precipitation data available from nearby Trappers Lake indicates that the amount of precipitation in May, 1995 was unusually high. Thus, the snowpack measurement of ^{35}S in June, 1995 may be higher than concentrations normally found at NWL for most years due to addition of ^{35}S from excess precipitation in May.

6. Sulphur-35 in Surface Waters

Concentrations in the spring water are much lower than the lake water, but generally above the detection limit, and show no consistent trend for either year. Sulphur-35 concentrations in spring water for both years average about $2 \pm 1 \text{ mBq}$

L⁻¹. The ³⁵S concentrations in the spring water are assumed to reflect concentrations found in shallow ground water. As the spring is located below the lakes, it may receive some inflow from the lakes through the fractures in the bedrock as well as general ground water flow. Sulphur-35 concentrations in spring water are substantially lower than concentrations found in the lakes in most cases, indicating that the sulphate and probably the water emerging from the spring are older than the lake waters. A simple piston-flow approach, using an initial concentration of 16 mBq L⁻¹ (average value for snowpack), gives an age estimate of about 8 months for the waters of this spring. A more realistic approach is to assume that the flow is a mixture of waters of various ages. Using a two member mixing model of recent snowmelt (³⁵S = 16 mBq L⁻¹ on 1 May) and 'old' water (³⁵S = 0 mBq L⁻¹) suggests that about 75% of the flow originates from older water. Thus, the majority of sulphate (and probably the water) in the spring is derived from precipitation prior to the previous winter. This is in accord with the observation that the sulphate concentrations in the spring do not correlate directly with those in precipitation.

Both lakes contain significant amounts of ³⁵S early in the summer in all years. UNWL had a larger initial concentration than NWL in 1995, although both had similar concentrations in 1996 (~8 mBq L⁻¹). In 1995 and 1996, ³⁵S concentrations, both as mBq L⁻¹ and mBq mg⁻¹SO₄, decrease in both lakes during the summer but increase in the autumn. In 1997, only NWL had a time series and the concentrations drop slightly in mBq L⁻¹ through the season. The ³⁵S data for the lakes indicate that a large fraction of the water and sulphate present at the beginning of the summer is derived from recent snowmelt. In all three years, ³⁵S concentrations are highest in the initial samples, usually collected in July. If the ³⁵S concentration was 20 mBq L⁻¹ for the snowpack in June 1995, simple decay would cause the concentration of the ³⁵S in lake water derived only from the snowpack to be slightly greater than 12 mBq L⁻¹ during the July sampling period. Concentrations for NWL and UNWL are 8.2 and 11.4 mBq L⁻¹, respectively, indicating that about 65% of the sulphate in NWL and 90% of the sulphate in UNWL was derived from melting snowpack that year. This assumes that the lake water had a concentration of 0 mBq L⁻¹ prior to snowmelt and very little ³⁵S was added after the beginning of June. (Rainfall also introduces ³⁵S which might tend to bias the results toward younger sulphate, so these results should be considered maximum estimates.) These values are high in 1995 due to the fact that the snowpack was larger than normal that year and melted late. For NWL, the 1996 results are similar to the 1995 results but for UNWL a lower fraction of new sulphate is indicated.

Flux calculations indicate that a large fraction of the ³⁵S present in UNWL in July is derived from the snowpack overlying the lake. Using an area of 0.25 ha, approximately a meter of water as snow lying directly above the lake, and a decay-corrected ³⁵S concentration of 12 mBq L⁻¹ in July, about 3x10⁴ Becquerels of ³⁵S can be derived directly from snow accumulated on the lake surface during winter. This accounts for all ³⁵S in UNWL in July of 1996 and about 75% present in July of 1995. As 1995 was a heavy snow year, the input from direct snow is probably

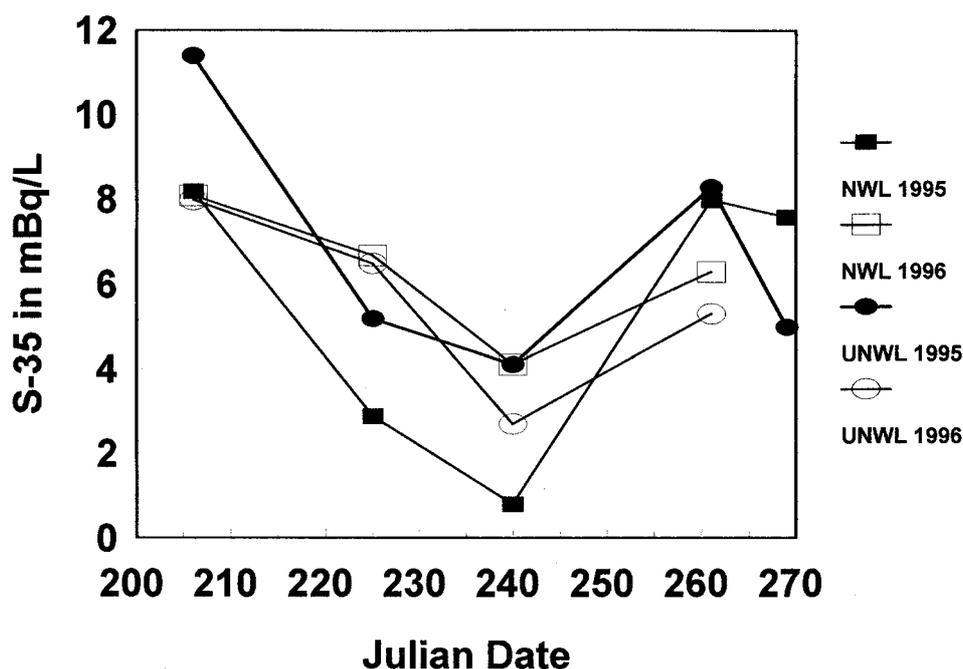


Figure 4. ^{35}S concentrations in mBq L^{-1} for Ned Wilson Lake (NWL) and Upper Ned Wilson Lake (UNWL) for 1995 and 1996. Timescale is given in Julian dates.

underestimated. For NWL, a direct input of ^{35}S in snow on the lakes surface of about 1.2×10^5 Becquerels is calculated, which represents about 25% of the ^{35}S present in the lake in July. Thus, about 75% of the ^{35}S (and sulphate) in the lake is derived from runoff.

The changes in ^{35}S (Figure 4) concentration during the summer reflect a complex interaction of radioactive decay, atmospheric deposition, and removal processes which are probably dominated by biogeochemical exchange. For 1995 and 1996, both NWL and UNWL show the same trends, with ^{35}S concentrations dropping, both in absolute terms and normalized to sulphate, during the summer until September when concentrations rise. The ^{35}S inventories present in the lakes during the summer can be compared to inventories expected if only decay affected the ^{35}S load. The initial ^{35}S inventory measured on the first sampling date of each year is used as the starting point, and inventories measured at later sampling dates are compared to it after the loss for decay is calculated. Figure 5 is a plot of the ratio of the ^{35}S inventory measured on a given sampling date divided by the ^{35}S inventory predicted from decay, versus Julian date for both lakes in 1995 and 1996 and NWL in 1997. Ratios less than one are an indication that ^{35}S is being removed from the system by processes other than decay. Ratios greater than one indicate that ^{35}S has been added to the lake since the initial spring melt.

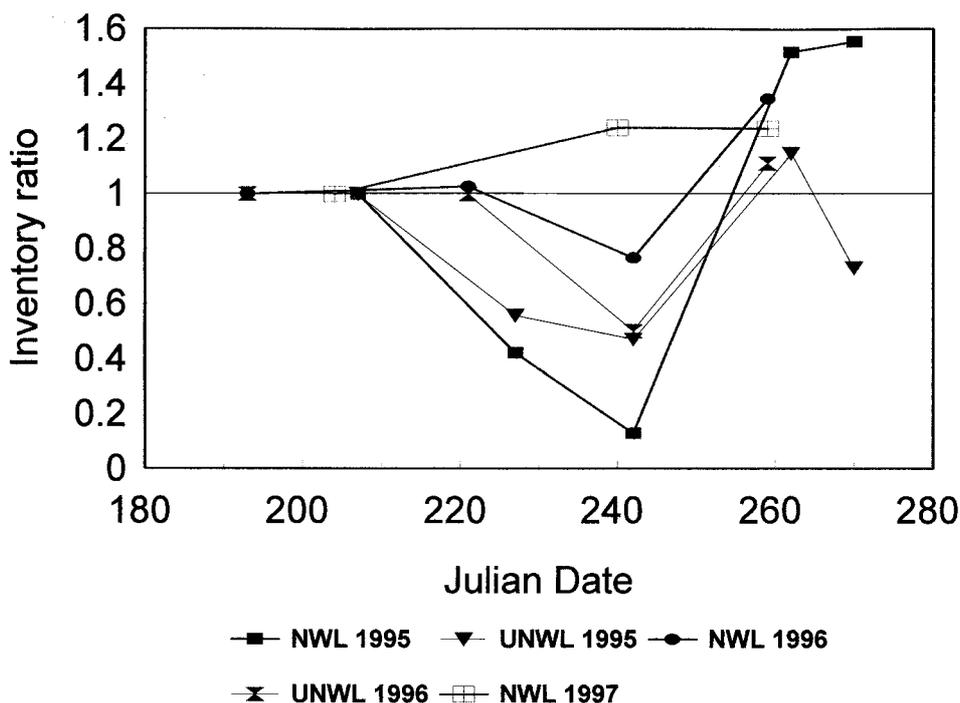


Figure 5. Ratio of measured ^{35}S inventories relative to inventories expected if only decay affected ^{35}S during the sampling season. Timescale is given in Julian dates. The initial inventories, measured on the first sampling date of the season, have a ratio of 1. Ratios less than one indicate that processes other than decay are removing ^{35}S . Ratios above 1 indicate that ^{35}S inventories are in excess of expected inventories indicating an addition of ^{35}S .

Figure 5 shows that there is a decrease in the inventories for some years in excess of that expected from decay. As neither sulphate nor ^{34}S shows any significant changes, Table II suggests that biogeochemical exchange is the only process that can explain the decrease in ^{35}S . Exchange has a minimal impact on the stable sulphur isotopic ratios, and if sulphate was returned to the lake, it would not result in a decrease in sulphate concentrations. Thus, the most likely scenario for the decrease of ^{35}S is biogeochemical cycling within the system, resulting in 'dead' sulphate replacing sulphate with ^{35}S in the water column.

The increases seen in both lakes in late summer can only be caused by precipitation bringing more recent ^{35}S into the system. At the Trappers Lake site, about 15% of the total annual precipitation occurred during the three summer sampling months (July–September) in 1995 while the corresponding percentage for 1996 was less than 10% (virtually no rain in August). However, as noted, the ^{35}S concentrations of rainfall tend to be much higher than snowpack concentrations. Rainfall also increased in September of 1995 and 1996 after relatively dry summers. The possibility of rain washing in ^{35}S that has accumulated as dryfall in the watershed

may also be an important factor. The larger increase seen in NWL compared to UNWL is likely due to small amounts of runoff which can also include any dry deposition occurring during the summer. In 1997, rainfall at Trappers Lake was much more evenly distributed during the summer with relatively high rainfall in August. Thus, the different pattern for ^{35}S in NWL (Figure 4) that year may be related to the more evenly spaced rainfall pattern in 1997.

7. Conclusions

Sulphur-35 has been used to determine which processes are important for sulphate cycling in the NWL watershed on seasonal timescales. It has shown that, when used in conjunction with sulphate and sulphur stable isotopic data, it can help determine which processes are important in small lakes, and give information on the rates of some processes. A small spring, representative of local shallow ground water, has low ^{35}S concentrations, indicating that it is composed of a mixture of recent snowmelt and precipitation that entered the watershed prior to the most recent winter. Concentrations of ^{35}S in the two lakes were very high in the initial sampling in July, indicating that a large fraction of the water in these lakes was derived from recent snowmelt. In UNWL, most of the recent water and sulphate could be accounted for by the melting of snow deposited on the lakes surface during the winter. In NWL, snow deposited on the lake could account for only 25% of the ^{35}S inventory, indicating that some runoff does occur from the surroundings. The change in ^{35}S concentrations in the two lakes during the summer showed a complex pattern, indicating that biogeochemical exchange was removing ^{35}S during the summer, and late summer precipitation added ^{35}S . The ^{35}S data does suggest that the chemistry in this watershed is sensitive to changes in sulphate deposition, which supports the earlier results showing that the response of lake water chemistry to changes in sulphate deposition is rapid (Campbell *et al.*, 1991).

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