

## RESEARCH ARTICLE

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## Key Points:

- Sulfur-35 is a unique tracer to quantify the Tibetan glacier melt
- Sulfur-35 vary significantly in aerosol, snow, and lake/river
- The age of Lake Nam Co surface water is ~10 months

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## Cosmogenic $^{35}\text{S}$ measurements in the Tibetan Plateau to quantify glacier snowmelt

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**Abstract** The cosmogenic radionuclide  $^{35}\text{S}$  ( $t_{1/2} \sim 87$  days) is a unique tracer for high-altitude air mass and has been used extensively to understand stratospheric air mass mixing. In this paper, we investigate if  $^{35}\text{S}$  can be utilized as an independent tracer to quantify glacier melt. We report the first measurements of  $^{35}\text{S}$  in samples collected from the Tibetan Plateau during 2009–2012 with an aim to interpret  $^{35}\text{S}$  in atmospheric particles and their deposition over glacier and snowmelts. Our measurements show that  $^{35}\text{S}$  activity in the aerosol phase varies from  $116 \pm 13$  to  $2229 \pm 52$  atoms/m<sup>3</sup> resulting in higher values during winter–spring and lower values during summer–autumn. This seasonality is likely due to higher mixing of  $^{35}\text{S}$ -rich stratospheric air masses during winter–spring and  $^{35}\text{S}$ -poor air masses from the Bay of Bengal during the Asian summer monsoon. The average  $^{35}\text{S}$  activity in the Zhadang glacier was found to be 3–8 times higher relative to the nearby lake water. The main source of  $^{35}\text{S}$  activity in the Zhadang glacier is atmospheric deposition, whereas both atmospheric deposition and glacier snowmelt are the primary sources in the Nam Co Lake. The focus of this study is to quantitatively determine the spatial and temporal variations in glacier snowmelt. In the future, extensive sampling of aerosols and snow is required for determining  $^{35}\text{S}$  in combination with stable oxygen isotopes in sulfate to better understand the glacier melt process and hydrological cycle on the Tibetan Plateau.

### 1. Introduction

The Tibetan Plateau (27°N–45°N, 70°E–105°E, average elevation ~4 km) is a home to nearly 36,800 glaciers amounting to a total glacial area of ~49,873 km<sup>2</sup> [Kang *et al.*, 2010]. It is the largest ice mass outside polar regions, and thereby known as the third pole. The ice melt from these glaciers sustains Asia's 10 major rivers including the Yangtze, Yellow, Mekong, Ganges, Brahmaputra, and the Indus Rivers, which are the sources of water for about 40% of the world's population, including China, India, and many Southeast and East Asian countries [Ding *et al.*, 2008; Xu *et al.*, 2009]. Recently, there has been evidence of climatic changes in the Tibetan Plateau, including increased warming, early snowmelt, and the retreat of high mountain glaciers [Duan and Wu, 2006; Kang *et al.*, 2010; Liu and Chen, 2000; Niu *et al.*, 2004; Wang *et al.*, 2008; Zhang *et al.*, 2012]. Satellite and field measurements show that the majority of glaciers on the Tibetan Plateau and the neighboring regions are shrinking more rapidly than previously thought [Ding *et al.*, 2006; Li *et al.*, 2008; Xiao *et al.*, 2007; Zhang *et al.*, 2012]. Local pollution and long-range transport of mineral dust and aerosols from the deserts of western China, Afghanistan/Pakistan, and the Middle East [Cong *et al.*, 2009; Fu *et al.*, 2006; Kang *et al.*, 2010; Lau *et al.*, 2006, 2010; Qian *et al.*, 2011; Wang *et al.*, 2008; Xu *et al.*, 2009, 2012] play an important role in temperature rise and albedo reduction, which consequently accelerates the melting of mountain glaciers. It has been proposed that atmospheric heating by Asian Brown Clouds [Ramanathan *et al.*, 2007] and snow-darkening effects by black carbon and dust [Qian *et al.*, 2011; Xu *et al.*, 2009] contribute substantially to the early snowmelt and retreat of mountain glaciers as well. Li *et al.* [2008] estimated that the glacier area might decrease by nearly 30% in 2050 with glacier runoff increasing to its maximum value in 2030. Tibetan glacier melt will have a severe impact on future water availability in Asian countries [Barnett *et al.*, 2005; Ding *et al.*, 2008; Immerzeel *et al.*, 2010]. Therefore, it is important to develop independent tracers to determine the extent of glacier melt and to verify model predictions in an effort to aid law regulations hoping to protect the

Tibetan glacier. In this paper, we demonstrate that the naturally occurring cosmogenic radioactive isotope of sulfur ( $^{35}\text{S}$ , half-life = 87 days) is a perfect tracer to understand hydrological processes on short time scales and also a unique tool to quantify the snowmelt on the Tibetan glacier, providing new information to enhance current model predictions.

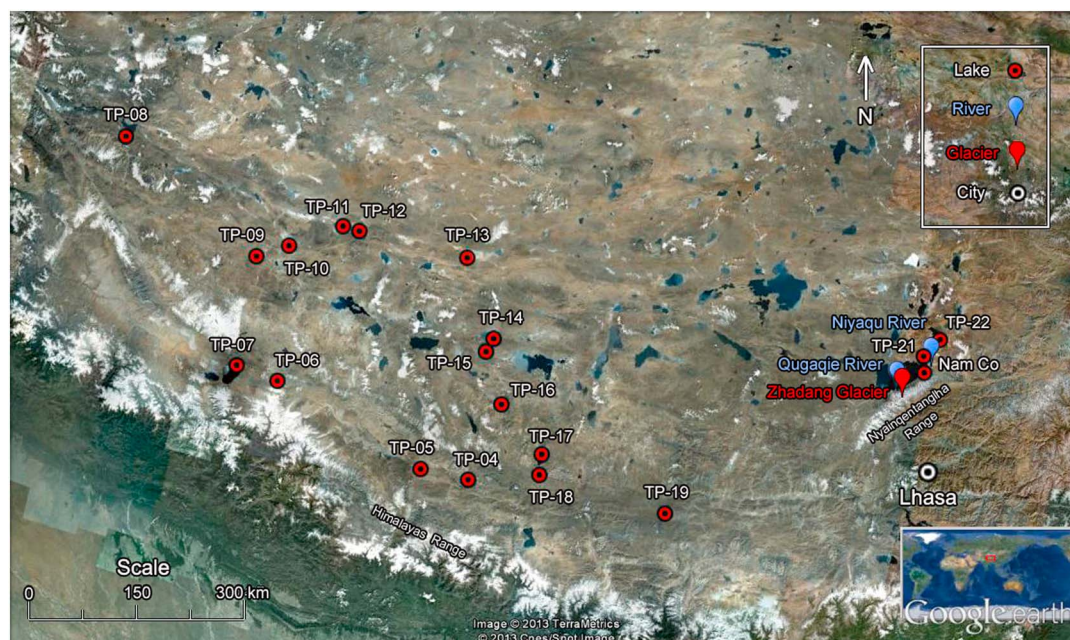
The cosmogenic radionuclide Sulfur-35 (half-life  $t_{1/2} = 87$  days) is produced by high-energy cosmic ray spallation of argon atoms ( $^{40}\text{Ar}$ ) in the atmosphere [Lal and Peters, 1967]. The production rate of  $^{35}\text{S}$  has both latitudinal and altitudinal dependence. For example, minimum production of  $^{35}\text{S}$  is seen in the equatorial troposphere, whereas maximum production occurs in the polar stratosphere. Following production,  $^{35}\text{S}$  rapidly oxidizes to  $^{35}\text{SO}_2$  (gas), which further gets oxidized to  $^{35}\text{SO}_4^{2-}$  particles. Due to higher  $^{35}\text{S}$  production rate and longer residence time in stratosphere, most  $^{35}\text{SO}_2$  and  $^{35}\text{SO}_4^{2-}$  produced in the stratosphere does not readily reach the boundary layer before decaying to  $^{35}\text{Cl}$ , except during stratospheric mixing events. Both  $^{35}\text{SO}_2$  and  $^{35}\text{SO}_4^{2-}$  are removed from the atmosphere by wet and dry deposition. At any given time,  $^{35}\text{S}$  activity in sulfate aerosols depends on several atmospheric parameters [Lal and Peters, 1967; Priyadarshi et al., 2011b, 2012; Tanaka and Turekian, 1991, 1995; Turekian and Tanaka, 1992]: (i) sampling location due to altitudinal and latitudinal dependency of  $^{35}\text{S}$  production rates, (ii) rate of oxidation of  $^{35}\text{S}$  to  $^{35}\text{SO}_4^{2-}$  via  $^{35}\text{SO}_2$ , set by oxidant levels of  $\text{OH}/\text{H}_2\text{O}_2/\text{O}_3$ , and UV photolysis flux, (iii) air mass mixing between the troposphere and stratosphere, which brings  $^{35}\text{S}$ -rich air mass from stratosphere to the boundary layer, and (iv) loss by scavenging processes (wet and dry deposition) which is controlled by wind speed, rain, and snow deposition rates. The residence time of  $^{35}\text{SO}_4^{2-}$  aerosol particles ( $<2\ \mu\text{m}$ ) in the Tibetan Plateau region (10–12 days) is less than its radioactive half-life (87 days). Therefore, the ultimate fate of  $^{35}\text{SO}_4^{2-}$  is largely deposition on snow and lake/river water. Since the atmospheric factors (altitude, wind speed, precipitation, and aerosol residence time) influencing  $^{35}\text{S}$  concentration is different at glaciers than other low-lying region, glacier snow is expected to contain significantly higher  $^{35}\text{S}$  activity as compared to lake and river water in the Tibetan Plateau.

The short half-life of  $^{35}\text{S}$  allows it to be used as a tracer of hydrological processes and sulfur migration between different reservoirs such as the atmosphere, hydrosphere, cryosphere, and biosphere that occur on time scales of months to approximately a year. In the past,  $^{35}\text{S}$  has been used as a tracer to understand the sulfate biogeochemical cycle and a dating tool for determining the source and age of water and quantitatively derive the contribution of glacier melt and ground water [Cecil et al., 1998; Cooper et al., 1991; Kester et al., 2003; Michel et al., 2001; Plummer et al., 2001]. For example, Cooper et al. [1991] used natural  $^{35}\text{S}$  along with stable sulfur isotopes to study spring runoff in a small Arctic watershed. Sueker et al. [1999] employed  $^{35}\text{S}$  technique to study the hydrology of three small basins in the Front Range of the Rocky Mountains.

The Tibetan Plateau contains closed catchments where glacier melts are stored in lakes through river runoff. In this closed system, monitoring  $^{35}\text{S}$  activity in atmospheric aerosols, glacier snow, glacier-fed lakes, and river water will enable us to determine the source and age of lake water developing a better understanding of the hydrological processes on the Tibetan Plateau. Here we present the first attempt to determine  $^{35}\text{S}$  in samples collected from various locations in the Tibetan Plateau.

## 2. Sampling

Atmospheric samples including aerosol,  $\text{SO}_2$ , glacier snow, lake, and river water were collected during 2009–2012 in the southern Tibetan Plateau to analyze total bulk sulfate concentration and  $^{35}\text{S}$  activity in  $^{35}\text{SO}_4^{2-}$  and  $^{35}\text{SO}_2$  phase. Aerosol and  $\text{SO}_2$  samples were collected at Nam Co station (N 30.77°, E 90.98°, 4730 m above sea level (asl)), which represents a clean continental background site for aerosol monitoring in Tibet [Cong et al., 2009]. This sampling station is established near Nam Co Lake, which is close to the Nyainqentanglha Mountain and about 50 km northeast of the Zhadang glacier. The bulk aerosol samples and  $\text{SO}_2$  samples were collected on quartz and KOH-treated quartz filter paper [Forrest and Newman, 1973], respectively. A high-volume aerosol sampler with a typical flow rate of  $\sim 1\ \text{m}^3\ \text{min}^{-1}$  was used to collect air sample continuously for 5 days. The sampling was done during each season; winter (December to February), spring (March to May), summer (June to September), and autumn (October to November). The



**Figure 1.** Details of the sampling locations in the Tibetan Plateau for  $^{35}\text{S}$  analysis in aerosol, glacier, and lake/river water sample collected during 2009–2012. Aerosol and glacier samples were collected at Nam Co station and Zhadang glacier, respectively. Water samples were collected from 19 different lakes and two rivers, as shown in the map.

annual average precipitation at Nam Co station is about 414 mm/yr mainly occurring between June and September under the influence of the Asian summer monsoon [Huang *et al.*, 2012; Kang *et al.*, 2011].

Water samples were collected from 19 different lakes and rivers (Qugaqie and Niyagu River), which are supported by winter–spring snowmelt, and summer precipitation mainly supports these rivers [Kang *et al.*, 2011]. Snow samples were collected from the Zhadang glacier on Mount Nyainqentanglha in the southern Tibetan Plateau (Figure 1 and Table 3). Fresh surface snow (0–24 cm) and deep snow (coarse-grained snow, 24–59 cm) samples were collected from the Zhadang glacier col (N 30.47°, E 90.65°, 5800 m) and Zhadang glacier terminal (N 30.48°, E 90.64°, 5500 m). At the Zhadang glacier terminal, surface and deep snow represents a depth of 0–11 cm and 11–20 cm, respectively. Three liters of lake and river water sample and snowmelt water were passed through an ion exchange (BioRad-Anion resin) resin column to trap sulfate ions. All resin-filled columns and aerosol filter papers were shipped to the University of California, San Diego, for the analysis of total sulfate concentration and radioactive  $^{35}\text{S}$  by following the standard chemical processing technique [Brothers *et al.*, 2010]. Organic contaminants and chlorine salts were removed before counting  $^{35}\text{S}$  activity in an ultralow-noise liquid scintillation counter [Brothers *et al.*, 2010; Priyadarshi *et al.*, 2011a]. The raw  $^{35}\text{S}$  data are corrected for the background activity and for the decay time between sampling and measurement.

### 3. Results and Discussion

For a quantitative understanding of temporal and spatial distribution of the Tibetan glacier melt, a well-defined mapping of  $^{35}\text{S}$  is required. The source of  $^{35}\text{S}$  in glacier snow is primarily atmospheric deposition of  $^{35}\text{SO}_2$  and  $^{35}\text{SO}_4^{2-}$ , whereas the sink of  $^{35}\text{S}$  is via physical loss of surface snow due to high wind, snowmelt, and by radioactive decay.

The  $^{35}\text{S}$  activity contained in lake and river water depends on the rate and source of snowmelt, atmospheric deposition of  $^{35}\text{SO}_2$  and  $^{35}\text{SO}_4^{2-}$ , hydrological properties such as the mixing rate of surface/deep water and on the biochemical sulfur cycle, which fractionates the sulfur reservoir. Since  $^{35}\text{S}$  chemically behaves similar to bulk (stable) sulfur, understanding  $^{35}\text{S}$  movement can be translated to the bulk sulfur cycle.

### 3.1. Variation in Bulk Sulfate Concentration

Bulk sulfate concentration in aerosol samples (44 samples), collected at Nam Co station during 2009–2012, varies from  $0.2 \text{ nmol/m}^3$  to  $11.2 \text{ nmol/m}^3$ , whereas  $\text{SO}_2$  concentration varies from  $0.2 \text{ nmol/m}^3$  to  $0.6 \text{ nmol/m}^3$  (Table 1). The annual mean concentration (mean  $\pm 1\sigma$ ) of  $\text{SO}_4^{2-}$  and  $\text{SO}_2$  was observed to be  $2.2 \pm 1.2 \text{ nmol/m}^3$  and  $0.4 \pm 0.1 \text{ nmol/m}^3$ , respectively (Table 2). Similar to bulk sulfate concentration (variation by a factor 10), the concentration of total suspended particle (TSP) at Nam Co station ranges from 48 to  $361 \text{ nmol/m}^3$  (annual average of  $67 \text{ nmol/m}^3$ ) with maximum concentrations observed during spring and summer periods [Cong *et al.*, 2009]. The average sulfate concentration at Nam Co station was higher as compared to Dome C in Antarctica [Priyadarshi *et al.*, 2011b] which probably due to local pollution as well as the long-range transport of mineral dust and aerosol from the desert in western China to Nam Co station [Li *et al.*, 2007].

The bulk sulfate concentration was higher in surface snow at the Zhadang glacier col as compared to the Zhadang glacier terminal (Table 3). Both at col and terminal, the deep snow samples showed a lower sulfate concentration relative to the surface snow samples. Bulk sulfate concentration in the Zhadang glacier range from  $0.4 \text{ }\mu\text{mol/L}$  to  $2.4 \text{ }\mu\text{mol/L}$  which is agreeing with measurements of bulk sulfate concentration in 2008 snow pit samples at Zhadang glacier col which vary from  $0.4 \text{ }\mu\text{mol/L}$  to  $11.7 \text{ }\mu\text{mol/L}$  [Huang *et al.*, 2012].

The bulk sulfate concentration in lake and river water varies from  $2.1 \text{ }\mu\text{mol/L}$  to  $578.1 \text{ }\mu\text{mol/L}$  and  $6.2 \text{ }\mu\text{mol/L}$  to  $71.4 \text{ }\mu\text{mol/L}$ , respectively (Table 3). The sulfate concentration in all lake and river water samples was higher by a factor of 2–500 and 6–70, respectively, compared to the average sulfate concentration measured in the Zhadang glacier ( $1.7 \text{ }\mu\text{mol/L}$ ). Wang *et al.* [2010] reported an average concentration of sulfate ion of  $2170 \text{ }\mu\text{mol/L}$  in Nam Co Lake water, whereas our data show  $359 \text{ }\mu\text{mol/L}$ . The difference in concentration might be due to the depth at which water samples were collected. Wang *et al.* [2010] collected samples at depths of 20 m to 68 m at an interval of 5 m whereas in our case, samples were collected at the surface level. The major sources of sulfate ion in the Nam Co Lake and river water are rock weathering, dissolution of evaporate minerals, and anthropogenic pollution [Wang *et al.*, 2010; Zhang *et al.*, 2008].

### 3.2. Variation in $^{35}\text{S}$ Activity in Atmospheric Aerosol and $\text{SO}_2$

Sulfur-35 activity in aerosol samples vary from  $241 \pm 6$  to  $2229 \pm 52$  atoms per  $\text{m}^3$  with an annual mean concentration of  $662 \pm 337$  (mean  $\pm 1\sigma$ ) atoms per  $\text{m}^3$ , whereas  $^{35}\text{SO}_2$  activity varies from  $45 \pm 7$  to  $189 \pm 9$  atoms per  $\text{m}^3$  with annual mean concentration  $118 \pm 44$  (mean  $\pm 1\sigma$ ) atoms per  $\text{m}^3$  (Table 1 and Figure 2). Sulfate-35 activity is expected to be higher than  $^{35}\text{SO}_2$  because the residence time of  $^{35}\text{SO}_4^{2-}$  in the atmosphere is longer (approximately weeks) [Priyadarshi *et al.*, 2011b] as compared to  $^{35}\text{SO}_2$  as it gets oxidized to  $^{35}\text{SO}_4^{2-}$  within a few days [Xu and Carmichael, 1997]. We compare the variation in background  $^{35}\text{S}$  activity measured at Nam Co with other sampling locations such as University of California, San Diego (UCSD)-Scripps pier, California, and Dome C, Antarctica. UCSD Scripps pier ( $32.85^\circ\text{N}$ ,  $117.28^\circ\text{W}$ , 10 m), located at sea level but at a similar latitude as Nam Co, observed lower  $^{35}\text{SO}_4^{2-}$  and  $^{35}\text{SO}_2$  activity ( $455 \pm 157$  ( $1\sigma$ ) and  $71 \pm 61$  ( $1\sigma$ ) atoms per  $\text{m}^3$ , respectively) relative to the Nam Co station [Priyadarshi *et al.*, 2012]. This is the result of the lower  $^{35}\text{S}$  production rate at sea level and lower probability of stratospheric intrusion events, bringing enriched  $^{35}\text{S}$  air mass to the surface. On the other hand,  $^{35}\text{SO}_4^{2-}$  activity at Dome C ( $74.3^\circ\text{S}$ ,  $123^\circ\text{E}$ , 3280 m) is higher compared to Nam Co, but surprisingly,  $^{35}\text{SO}_2$  activity at Dome C is comparative to Nam Co measurements. The natural background of  $^{35}\text{SO}_4^{2-}$  and  $^{35}\text{SO}_2$  at Dome C, Antarctica during summer is 728 and 189 atoms per  $\text{m}^3$ , respectively [Hill-Falkenthal *et al.*, 2013; Priyadarshi *et al.*, 2011b]. Dome C is situated at a similar altitude as Nam Co; however, it has the highest  $^{35}\text{S}$  production rate due to the higher abundance of cosmic rays at the pole.

The monthly average activities of  $^{35}\text{SO}_4^{2-}$  at Nam Co exhibit higher activity during winter–spring (December to May) relative to summer–autumn (July to November) (Table 2). The  $^{35}\text{SO}_4^{2-}$  activity ( $2229 \pm 52$  ( $1\sigma$ ) atoms per  $\text{m}^3$ ) measured on 1 March 2012 is the highest activity measured so far in a natural atmospheric sample. This result is influenced by an influx of stratospheric air mass enriched in  $^{35}\text{S}$  intruding into the boundary layer. The stratospheric intrusion events occur more frequently during the winter [Chen *et al.*, 2011]. The positioning of Tibetan Plateau (mainly high altitude) on the world map is such that it serves as a gateway for tropospheric-stratospheric air mass exchange [Fu *et al.*, 2006; Park *et al.*, 2009], which enhances the probability of downward stratospheric air masses mixing. During stratospheric-tropospheric exchange, a

**Table 1.** Sulfur-35 Activity in Aerosol and SO<sub>2</sub> Collected at Nam Co Station<sup>a</sup>

Sample Id	Collection Date	Julian Dates	Sample Volume (in m <sup>3</sup> )	Total DPM	[SO <sub>4</sub> ] (nmol/m <sup>3</sup> )	<sup>35</sup> S (atoms per m <sup>3</sup> )
			<i>SO<sub>4</sub><sup>2-</sup> Aerosol</i>			
TSP-T1	10/30/2010	10303	1414	0.91	1.0	116 ± 13
TSP-T2	11/25/2010	10329	4969	8.9	—	324 ± 6
TSP-T3	12/7/2010	10341	4331	44.7	—	1872 ± 9
TSP-T4	2/7/2011	11038	5344	26.5	—	899 ± 4
TSP-T5	2/14/2011	11045	6076	20.5	0.3	611 ± 8
TSP-T6	2/21/2011	11052	6455	11.5	0.2	323 ± 6
TSP-T7	2/27/2011	11058	6366	25.9	0.4	739 ± 7
TSP-T8	3/20/2011	11079	7790	16.3	0.5	379 ± 10
TSP-T9	3/25/2011	11084	6451	26.7	2.2	750 ± 12
TSP-T10	3/31/2011	11090	6526	12.3	0.3	342 ± 10
TSP-T11	4/10/2011	11100	4269	29.3	2.0	1246 ± 16
TSP-T12	4/22/2011	11112	4564	27.6	1.1	1098 ± 17
TSP-T13	4/30/2011	11120	6518	13.4	0.6	372 ± 10
TSP-T14	5/06/2011	11126	6886	10.7	2.0	281 ± 9
TSP-T15	5/17/2011	11137	9081	24.2	1.0	483 ± 7
TSP-T16	5/23/2011	11143	6401	20.9	1.9	593 ± 9
TSP-T17	6/1/2011	11152	11050	19.5	1.1	319 ± 5
TSP-T18	12/3/2011	11337	6784	17.9	4.5	478 ± 10
TSP-T19	12/18/2011	11352	6699	23.1	4.1	625 ± 9
TSP-T20	12/24/2011	11358	6700	25.9	1.0	701 ± 9
TSP-T21	12/30/2011	11364	5481	15.1	1.7	501 ± 9
TSP-T22	1/12/2012	12012	4300	19.7	6.3	831 ± 10
TSP-T23	1/15/2012	12015	4195	19.8	5.0	858 ± 30
TSP-T24	1/23/2012	12023	4152	18.6	6.0	814 ± 29
TSP-T25	1/29/2012	12029	4059	17.2	2.4	813 ± 28
TSP-T26	3/08/2012	12068	4147	51.0	3.5	2229 ± 52
TSP-T27	3/22/2012	12082	4247	27.2	3.1	1164 ± 26
TSP-T28	4/28/2012	12119	4233	37.8	7.4	1623 ± 41
TSP-T29	5/4/2012	12125	5584	52.0	6.7	1690 ± 29
TSP-T30	5/8/2012	12129	5778	27.8	11.2	874 ± 27
TSP-T31	5/18/2012	12139	7094	52.4	6.1	1339 ± 13
TSP-T32	5/22/2012	12143	6276	35.4	4.1	1024 ± 14
TSP-T33	5/26/2012	12147	5740	37.9	4.4	1197 ± 14
TSP-T34	7/3/2012	12185	7188	9.5	0.8	241 ± 6
TSP-T35	7/7/2012	12189	5566	11.5	—	376 ± 9
TSP-T36	7/12/2012	12194	7206	11.5	0.5	290 ± 6
TSP-T37	7/16/2012	12198	5762	8.1	0.6	256 ± 7
TSP-T38	7/24/2012	12206	10161	28.1	1.0	502 ± 5
TSP-T39	7/29/2012	12211	7069	5.1	0.4	145 ± 5
TSP-T40	8/2/2012	12215	6074	30.8	1.6	920 ± 7
			<i>SO<sub>2</sub> Gas</i>			
SO <sub>2</sub> -T1	11/25/2010	10329	4969	3.29	—	120 ± 8
SO <sub>2</sub> -T2	12/7/2010	10341	4331	4.52	—	189 ± 9
SO <sub>2</sub> -T3	2/7/2011	11038	5344	4.65	—	158 ± 5
SO <sub>2</sub> -T4	3/20/2011	11079	7790	3.90	0.4	91 ± 10
SO <sub>2</sub> -T5	3/25/2011	11084	6451	4.24	0.5	119 ± 11
SO <sub>2</sub> -T6	3/31/2011	11090	6526	5.88	0.4	163 ± 11
SO <sub>2</sub> -T7	4/10/2011	11100	4269	4.03	0.6	171 ± 15
SO <sub>2</sub> -T8	4/22/2011	11112	4564	3.78	0.4	150 ± 16
SO <sub>2</sub> -T9	4/30/2011	11120	6518	4.27	0.2	119 ± 11
SO <sub>2</sub> -T10	5/17/2011	11137	9081	4.56	0.2	91 ± 7
SO <sub>2</sub> -T11	5/23/2011	11143	6401	1.61	—	45 ± 8
SO <sub>2</sub> -T12	6/1/2011	11152	11050	4.22	—	69 ± 5
SO <sub>2</sub> -T13	12/3/2011	11337	6784	1.70	—	46 ± 7
SO <sub>2</sub> -T14	12/24/2011	11358	6700	4.75	0.3	129 ± 6
SO <sub>2</sub> -T15	12/30/2011	11364	5481	3.33	0.3	110 ± 7

<sup>a</sup>A higher <sup>35</sup>SO<sub>4</sub><sup>2-</sup> activity in winter–spring is due to higher influx of stratospheric air mass into the boundary layer, whereas lower <sup>35</sup>SO<sub>4</sub><sup>2-</sup> activity in summer is due to a well-mixed boundary layer and the onset of Asian summer monsoon, which brings <sup>35</sup>S-poor air from Bay of Bengal to the Tibetan Plateau. DPM = disintegrations per minute.

**Table 2.** Monthly Average Concentration of Bulk Sulfate and  $^{35}\text{SO}_4^{2-}$  Activity in Aerosol Collected at Nam Co Station<sup>a</sup>

Month of the Year <sup>b</sup>	Average $\text{SO}_4^{2-}$ Concentration (nmol/m <sup>3</sup> ) (Mean $\pm 1\sigma$ )	Average $^{35}\text{SO}_4^{2-}$ Concentration (atoms/m <sup>3</sup> ) (Mean $\pm 1\sigma$ )
January (4)	5.0 $\pm$ 1.8	829 $\pm$ 21
February (4)	0.3 $\pm$ 0.1	643 $\pm$ 244
March (5)	1.9 $\pm$ 1.5	973 $\pm$ 777
April (4)	2.8 $\pm$ 3.2	1085 $\pm$ 524
May (8)	4.7 $\pm$ 3.3	935 $\pm$ 472
June (1)	1.1	319
July (6)	0.6 $\pm$ 0.2	302 $\pm$ 123
August (1)	1.6	920
September (0)	— <sup>c</sup>	— <sup>c</sup>
October (1)	1.0	116
November (1)	— <sup>c</sup>	324
December (5)	— <sup>c</sup>	835 $\pm$ 586

<sup>a</sup>Winter–spring shows higher concentration of  $^{35}\text{SO}_4^{2-}$  whereas summer–autumn has the lowest. Bulk sulfate concentration does not exhibit any seasonality.

<sup>b</sup>Number of samples analyzed during 2010–2012 is given within the parenthesis.

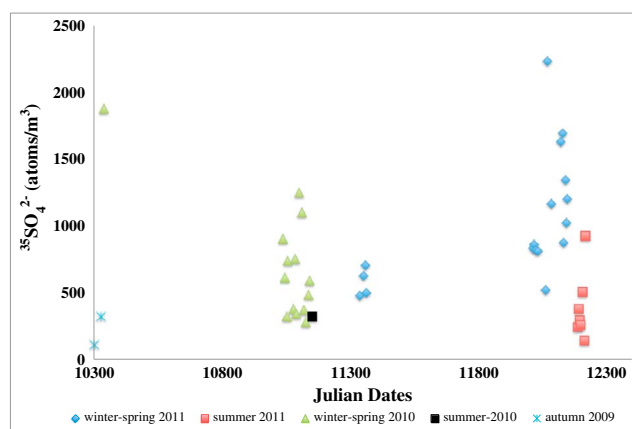
<sup>c</sup>No data available.

**Table 3.** Sulfate-35 Measured in Lake and River Water Collected From the Southern Tibetan Plateau

Sample Id	Sample Location (Elevation in m asl)	Collection Date	Sample Volume	$[\text{SO}_4^{2-}]$ (μmol/L)	DPM	$^{35}\text{S}$ (atoms per L) <sup>a</sup> 10 <sup>3</sup>
<i>Lake Water</i>						
TP-04	N 29.58°, E 84.77°, 4630	11/12/2009	3	7.2	0.24	14 $\pm$ 5
TP-05	N 29.69°, E 84.13°, 4570	11/12/2009	3	12.4	0.33	20 $\pm$ 6
TP-06	N 30.65°, E 82.15°, 4790	11/12/2009	3	205.4	0.53	32 $\pm$ 6
TP-07 <sup>b</sup>	N 30.81°, E 81.58°, 4610	11/14/2009	3	2.1	0.16	20 $\pm$ 5
TP-08	N 33.43°, E 79.78°, 4260	11/15/2009	3	287.3	0.22	13 $\pm$ 5
TP-09	N 32.09°, E 81.78°, 4640	11/16/2009	3	229.4	0.72	43 $\pm$ 6
TP-10	N 32.23°, E 82.23°, 4410	11/16/2009	3	63.1	0.17	10 $\pm$ 5
TP-11	N 32.48°, E 82.97°, 4410	11/16/2009	3	179.9	0.33	20 $\pm$ 6
TP-12	N 32.43°, E 83.2°, 4440	11/16/2009	3	369.1	0.46	28 $\pm$ 5
TP-13	N 32.15°, E 84.7°, 4400	11/17/2009	3	222.5	0.46	28 $\pm$ 6
TP-14 <sup>b</sup>	N 31.22°, E 85.08°, 4650	11/17/2009	3	464.9	0.30	22 $\pm$ 5
TP-15	N 31.07°, E 84.98°, 4760	11/17/2009	3	190.6	0.60	36 $\pm$ 5
TP-16	N 30.46°, E 85.2°, 4860	11/18/2009	3	287.5	1.56	94 $\pm$ 6
TP-17	N 29.89°, E 85.75°, 5170	11/18/2009	3	578.1	0.60	36 $\pm$ 6
TP-18	N 29.65°, E 85.72°, 5080	11/18/2009	3	200.9	0.36	22 $\pm$ 5
TP-19	N 29.20°, E 87.4°, 4290	11/18/2009	3	82.5	0.39	23 $\pm$ 6
TP-21	N 31.15°, E 91.22°, 4671	9/15/2010	3	150.2	0.32	19 $\pm$ 6
TP-22	N 30.95°, E 90.97°, 4746	9/15/2010	3	333.4	0.52	32 $\pm$ 6
Nam Co Lake	N 30.52°, E 90.98°, 4730	9/7/2010	3	359.3	0.31	19 $\pm$ 6
Nam Co Lake	N 30.52°, E 90.98°, 4730	12/16/2010	3	—	0.40	24 $\pm$ 7
Nam Co St	N 30.77°, E 90.98°, 4730	9/7/2010	3	394.4	0.34	21 $\pm$ 6
Nam Co St	N 30.77°, E 90.98°, 4730	11/19/2009	3	566.7	0.48	29 $\pm$ 6
Nam Co St	N 30.77°, E 90.98°, 4730	12/16/2010	3	—	0.28	19 $\pm$ 6
Miramar Lake <sup>a</sup>	N 32.92°, W 117.09°, 10	1/31/2010	4	257.2	0.17	8 $\pm$ 4
Tahoe Lake <sup>a</sup>	N39.08°, W 120.03°, 1897	11/10/2010	3	—	0.44	26 $\pm$ 7
<i>Zhadang Glacier Col</i>						
Surface snow	N 30.47°, E 90.65°, 5800	9/10/2010	4.38	2.4	2.33	97 $\pm$ 5
Deep snow	N 30.47°, E 90.65°, 5800	9/10/2010	3.42	1.0	0.77	41 $\pm$ 6
<i>Zhadang Glacier Terminal</i>						
Surface snow	N 30.48°, E 90.64°, 5500	9/10/2010	4.45	0.9	1.57	64 $\pm$ 5
Deep snow	N 30.48°, E 90.64°, 5500	9/10/2010	5.4	0.4	0.75	25 $\pm$ 4
<i>River</i>						
Quagaqie River	N 30.57°, E 90.57°, 4800	9/10/2010	3	71.4	0.30	18 $\pm$ 6
Niyaqu River	N 30.83°, E 91.06°, 4750	9/7/2010	3	6.2	0.27	16 $\pm$ 6
Niyaqu River	N 30.83°, E 91.06°, 4750	12/16/2010	3	—	0.25	15 $\pm$ 5

<sup>a</sup>Samples are collected from California and Sierra Nevada, United States.

<sup>b</sup>Half water sample was lost from TP07 and one fifth of water sample was lost from TP14. Sulfur-35 activities were corrected accordingly.



**Figure 2.** Variation in  $^{35}\text{S}$  activity in sulfate aerosols collected at Nam Co station during 2010 to 2012. Higher concentration of  $^{35}\text{SO}_4^{2-}$  during winter–spring (December to May) is due to stratospheric air mass ( $^{35}\text{S}$ -rich) influx into the boundary layer. Mixing of  $^{35}\text{S}$ -poor air mass originated from Bay of Bengal during Indian summer monsoon decreases the  $^{35}\text{SO}_4^{2-}$  activity in summer.

Plateau is expected to lie within this range. A nearly 5% mixing of stratospheric air mass in the boundary layer explains the increase in  $^{35}\text{S}$  concentration observed on 1 Mar 2012.

A relatively lower concentration of  $^{35}\text{SO}_4^{2-}$  observed during summer at Nam Co is mainly due to scavenging of  $^{35}\text{SO}_4^{2-}$  by heavy summer monsoon precipitation. The southern part of the Tibetan Plateau is influenced by the Asian summer monsoons from June to September. In addition to the frequent precipitation (which removes  $^{35}\text{SO}_4^{2-}$  particles considerably from the atmosphere), the Asian monsoon air masses also dilutes the  $^{35}\text{SO}_4^{2-}$  and  $^{35}\text{SO}_2$  concentrations at Nam Co, because the monsoon air mass originates at a lower altitude (sea level at Bay of Bengal) [Liu *et al.*, 2009] containing low  $^{35}\text{S}$  concentration. Moreover,  $^{35}\text{SO}_2$  concentrations during summer are expected to be low because of the increased oxidation rate of  $^{35}\text{SO}_2$  to  $^{35}\text{SO}_4^{2-}$  by atmospheric oxidants, including  $\text{OH}/\text{O}_3/\text{H}_2\text{O}_2$  [Alexander *et al.*, 2005; Savarino *et al.*, 2000]. Similar to  $^{35}\text{S}$ , summer monsoon air masses are also devoid of ozone and are held responsible for the loss of ozone observed in the Tibetan atmosphere during summer when ozone poor air masses from the Bay of Bengal ascends toward the midtroposphere (8–12 km) leading to a decrease in ozone concentration [Tobo *et al.*, 2008; Zhou and Zhang, 2005; Zou, 1996]. Additionally, the strong surface heating during summer leads to a well-mixed boundary layer, up to 8 km asl [Yang *et al.*, 2004] and increases the height of the troposphere up to ~17 km asl [Fu *et al.*, 2006; Tian *et al.*, 2008] leading to lower occurrence of stratospheric intrusion events. Therefore, a frequent precipitation and a decrease in stratospheric intrusion events are mainly responsible for the decrease in  $^{35}\text{S}$  activity during summer.

### 3.3. Sulfate-35 Activity in Zhadang Glacier and Lake/River Water

The average  $^{35}\text{SO}_4^{2-}$  activity in Zhadang glacier snow was measured to be  $57 \pm 31 \times 10^3$  (mean  $\pm 1\sigma$ ) atoms per L, with a range of  $25 \pm 4 \times 10^3$  to  $97 \pm 5 \times 10^3$  atoms per L. Surface snow collected at the Zhadang glacier terminal shows the lowest  $^{35}\text{SO}_4^{2-}$  activity ( $64 \times 10^3$  atoms/L) whereas Zhadang glacier col sample has the highest activity ( $97 \times 10^3$  atoms/L). The  $^{35}\text{SO}_4^{2-}$  activity in the deep snow (coarse grained) was found to be 3–4 times lower than the surface snow and is probably due to the loss of  $^{35}\text{SO}_4^{2-}$  by ion percolation and radioactive decay over time. Sulfate-35 activity in lake water varies from  $10 \pm 5 \times 10^3$  to  $94 \pm 6 \times 10^3$  atoms per L (Table 3) and was found to be lower by 3–8 times as compared to  $^{35}\text{SO}_4^{2-}$  in Zhadang glacier. No correlation between  $^{35}\text{S}$  activity and bulk sulfate concentrations with the lake size or altitude was observed.

We compared  $^{35}\text{S}$  activity measured at the Tibetan lakes with two other lakes in California: Miramar Lake [N 32.92°, W 117.09°, 10 m] and Lake Tahoe [N 39.08°, W 120.03°, 1897 m]. Miramar Lake is a fresh water lake, situated at sea level (low- $^{35}\text{S}$  production rate), and exhibits a lower  $^{35}\text{SO}_4^{2-}$  concentration,  $8 \pm 4 \times 10^3$  atoms per L (Table 3). On the other hand,  $^{35}\text{SO}_4^{2-}$  activity in Lake Tahoe, (situated in the California-Sierra Nevada

fraction of stratospheric air mass containing higher concentrations of  $^{35}\text{SO}_4^{2-}$  and  $^{35}\text{SO}_2$ , mixes directly into the boundary layer directly (deep mixing) or indirectly after a time lag (shallow mixing) and causes an enhancement in  $^{35}\text{SO}_4^{2-}$  concentrations in the boundary layer. Since the  $^{35}\text{S}$  activity has not been measured directly, a mathematical model based on  $^{35}\text{S}$  production rate, atmospheric chemistry, and air mass mixing has been used. The model calculation predicts that the  $^{35}\text{S}$  activity in the lower stratosphere is 6316 atoms per  $\text{m}^3$  and 13,383 atoms per  $\text{m}^3$  at Scripps pier and Dome C, respectively [Priyadarshi *et al.*, 2011b, 2012], demonstrating the nonuniformity of  $^{35}\text{S}$  concentration in the stratosphere. The stratospheric  $^{35}\text{S}$  activity at the Tibetan

region, at an altitude of  $\sim 2$  km) was measured to be  $26 \pm 7 \times 10^3$  atoms per L, which is 3 times higher than the activity observed in Miramar Lake. Similar to the Tibetan lakes, Lake Tahoe is a snow fed lake and receives higher  $^{35}\text{S}$  flux by atmospheric deposition; thereby,  $^{35}\text{SO}_4^{2-}$  activity in Lake Tahoe is similar to the activity measured in Tibetan lakes.

### 3.4. Age Determination Based on $^{35}\text{S}$

At Zhadang glacier, snowmelt is strong during summer; thereby, snow stored as ice is negligible which often leads to a negative or deficit in annual mass balance [Zhang *et al.*, 2013]. Nearly all of the snowpack melts during summer and flows into runoff whereas snow sublimation is weak during rest of the year. Since the Tibetan lakes and rivers are mainly derived from snowmelt,  $^{35}\text{SO}_4^{2-}$  activity in lake-river water is expected to be the same as the  $^{35}\text{SO}_4^{2-}$  activity in Zhadang glacier if the source of water is exclusively derived from the Zhadang glacier. However, our measurements show that the average  $^{35}\text{SO}_4^{2-}$  concentration in the Nam Co Lake surface water is approximately 4–7 times lower compared to the Zhadang glacier. The possible reasons are the following: (1) Zhadang glacier may not be the only source of input water to the lake [Zhang *et al.*, 2013]. Probably, the snowmelt of both fresh and old ice (containing less or no  $^{35}\text{S}$  activity) of different glacier contributes significantly toward the lake water. (2) Lake Nam Co is a well-mixed lake where deep water containing no  $^{35}\text{S}$  from the lake bottom mixes with the surface water and consequently dilutes the surface water  $^{35}\text{S}$  activity. Considering the low surface temperature and the size of the Nam Co Lake, the second biggest lake on the Tibetan Plateau (total area 2017 km<sup>2</sup> and total catchment area of 10,680 km<sup>2</sup>, average depth  $\sim 90$  m), it is unlikely that the lake is well mixed [Zhou *et al.*, 2013]. Our  $^{35}\text{S}$  data indicate the existence of multiple water sources from the Nam Co Lake, which agrees with the estimation from Zhou *et al.* [2013] that precipitation and meltwater runoff from the glaciers account for 23–28% and 7–22% of the total water input to the lake, respectively.

The  $^{35}\text{SO}_4^{2-}$  activities in the Qugaqie and Niyaqu rivers (the closest rivers to the Zhadang glacier) were nearly the same ( $\sim 18 \times 10^3$  atoms per L) and found to be 6–9 times lower than the activity measured at the Zhadang glacier. Interestingly, the  $^{35}\text{S}$  activity measured in the Niyaqu River during September and December 2010 shows a similar value, indicating a steady state equilibrium between the input and output fluxes, whereas  $^{35}\text{S}$  activity at Lake Nam Co is 30% lower during September 2010 compared to December 2010. This likely results from either (or both) higher biological uptake rate of  $^{35}\text{S}$  by lake biota during summer (September) or less atmospheric deposition of  $^{35}\text{S}$  by dry or wet processes due to significantly lower  $^{35}\text{S}$  activity in atmosphere (as discussed in section 3.2) caused by several precipitations during Indian summer monsoon.

We note that the bulk sulfate concentrations and  $^{35}\text{SO}_4^{2-}$  activities vary considerably among the Tibetan lakes and rivers. This variation in  $^{35}\text{S}$  activities is due to changes in the relative contribution of different glacier sources, snowmelt of fresh snow versus older ice, difference in the onset and duration of snowmelt, and deep water mixing which consequently result in mixing of water-soluble sulfate containing variable  $^{35}\text{S}$  activity corresponding to different ages. There are several other processes that can decrease the  $^{35}\text{SO}_4^{2-}$  concentration in lake water such as biogeochemical exchange and biological uptake/reduction [Michel *et al.*, 2001]. However, these processes will not fractionate between  $^{35}\text{SO}_4^{2-}$  and bulk sulfate concentrations as radioactive  $^{35}\text{S}$  behaves exactly similar to stable sulfate. In contrast, mineral weathering serves as a source of bulk sulfate devoid of radiogenic  $^{35}\text{SO}_4^{2-}$  due to the short half-life of  $^{35}\text{S}$  and decreases the overall specific activity (defined as number of  $^{35}\text{S}$  atoms per mole of bulk sulfate). Based on our limited data set, it is not possible to understand the biochemical cycle of  $^{35}\text{S}$  in lake/river water and estimate the  $^{35}\text{S}$  loss (dilution) due to biological uptake (mineral weathering) quantitatively. A larger data set with increased sample resolution can potentially yield constraints on biological uptake and weathering processes.

Assuming the Zhadang glacier as the main contributor of snowmelt/runoff water to the Nam Co Lake and no loss of  $^{35}\text{S}$  other than radioactive decay loss, we estimate the age of surface lake water.

The  $^{35}\text{SO}_4^{2-}$  activity in the Zhadang glacier was measured during late summer (September) when a fraction of fresh snow has already melted. Summertime  $^{35}\text{S}$  activity in snow is not the same as the  $^{35}\text{S}$  activity in springtime (before the onset of snowmelt), because of the loss of snow as well as in situ radioactive decay. We estimated springtime (May)  $^{35}\text{SO}_4^{2-}$  activity in surface snow to be  $214 \pm 62 \times 10^3$  (mean  $\pm 1\sigma$ ) atoms per L by correcting the late summertime (September)  $^{35}\text{SO}_4^{2-}$  activity (Avg =  $80 \pm 23 \times 10^3$  atoms per L) for in situ



radioactive decay. Utilizing the average  $^{35}\text{SO}_4^{2-}$  activities for the Zhadang glacier and lake water during spring and summer, respectively, the average age of the Nam Co Lake surface water and Qugaqie and Niyaqu rivers are estimated to be  $9.7 \pm 0.7$  months and  $10.8 \pm 0.4$  months, respectively, which is an upper limit of the estimate. This is the age of the Nam Co Lake surface water and is order of magnitude younger than the reservoir age. Based on radioactive isotope ( $^{137}\text{Cs}$  and  $^{210}\text{Pb}$ ) tracer technique, the reservoir age of the sediment core from the Nam Co Lake was estimated to vary from 2476 to 1200 years [Zhu *et al.*, 2008]. Considering other losses of  $^{35}\text{S}$ , such as biological uptake, deep water or ground water mixing (with no  $^{35}\text{S}$ ) will lower the estimated age. The advantage of  $^{35}\text{S}$  is that it determines the age of surface water very accurately compared to other radioactive isotopes ( $^{137}\text{Cs}$ ,  $^{210}\text{Pb}$ , and  $^{14}\text{C}$ ) due to its short half-life (83 days) and thus helps to understand the seasonal pattern of the Tibetan glacier melt.

#### 4. Conclusion and Future Scope of the Present Work

Sulfur-35 activity was measured in atmospheric samples collected from the Tibetan Plateau. Sulfur-35 in sulfate aerosols varies from 116 to 2229 atoms per  $\text{m}^3$  with higher activities during winter–spring and lower activities during summer–autumn. Frequent stratospheric intrusion events, which bring  $^{35}\text{S}$  rich stratospheric air mass into the boundary layer leads to a higher  $^{35}\text{S}$  activity during winter. Sulfate-35-poor air mass coming from the Bay of Bengal during the Asian summer monsoon season results in lower  $^{35}\text{S}$  activity during summer. The average concentration of  $^{35}\text{SO}_4^{2-}$  in the Zhadang glacier surface snow is  $80 \pm 23 \times 10^3$  atoms per L and is 3–8 times higher than the  $^{35}\text{SO}_4^{2-}$  activities measured in lake and river water. Based on our limited data set and neglecting deep water mixing and the biological sulfur uptake, the average age of surface lake water was determined to be  $10 \pm 1$  months. In the future, an extensive sampling with high temporal resolution measuring  $^{35}\text{S}$  and the oxygen isotopic anomaly in sulfate would lead to better constraints on the hydrological cycle and help quantitative understanding of glacier melt at the Tibetan Plateau.

The radiogenic  $^{35}\text{S}$  provides an ideal clock to understand the natural sources and sinks of water-soluble sulfate. It is utilized to understand the movement between different sulfur reservoirs and ultimately to estimate the percentages of glacier melt. Careful analysis of  $^{35}\text{SO}_4^{2-}$  concentrations in glacier and lake-river water throughout the year, particularly before the onset of snowmelt and during peak discharge time periods will help better constrain the contribution of glacier snowmelt as an input flux. Triple oxygen isotope abundances in sulfate provide additional information regarding  $^{35}\text{SO}_2$  oxidation pathways and the presence of the oxidant ( $\text{O}_3$ ,  $\text{H}_2\text{O}_2$ , and  $\text{OH}$ ) levels in the atmosphere [Alexander *et al.*, 2005; Savarino *et al.*, 2000]. The oxygen anomaly ( $\Delta^{17}\text{O}$ ) in sulfate is created by mass independent chemical processes and is observed only in the atmospherically derived sulfate. In the future, a more finessed technique involving both  $^{35}\text{S}$  and  $\Delta^{17}\text{O}$  anomaly in sulfate [Hill-Falkenthal *et al.*, 2012; Lee and Thiemens, 2001] will be used to understand the sulfate biogeochemical cycle and to determine the unaltered fraction of atmospherically derived sulfate in the lake-river water.

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