A 1000 year history of atmospheric sulfate concentrations in southern Asia as recorded by a Himalayan ice core

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[1] A sulfate record covering the period A.D. 1000–1997 from the Dasuopu glacier in the Himalayas reveals that this site is sensitive to anthropogenic activity originating in southern Asia. Prior to 1870 atmospheric sulfate concentrations were relatively low and constant, but thereafter concentrations have increased and since 1930 the rate of increase has accelerated rapidly. This accelerating trend in sulfate deposition is paralleled by growing SO_2 emissions over southern Asia resulting from the increased energy demand. The concentration of sulfate deposited in the last 50 years exceeds that for any prior 50-year period in the last millennium. Unlike the Greenland ice core-derived sulfate concentrations that have declined since the 1970s, sulfate concentrations deposited on the Himalayan ice fields continue to increase, having nearly doubled since 1970. This reflects regional differences between Europe and Asia in source strength and transport pathways for atmospheric sulfate, as well as differing degrees of environmental regulation. **Citation:** Duan, K., L. G. Thompson, T. Yao, M. E. Davis, and E. Mosley-Thompson (2007), A 1000 year history of atmospheric sulfate concentrations in southern Asia as recorded by a Himalayan ice core, *Geophys. Res. Lett.*, *34*, L01810, doi:10.1029/2006GL027456.

1. Introduction

[2] Sulfate aerosols cause climate forcing by reflecting and absorbing radiation, as well as through indirect effects that alter cloud cover and cloud albedo [*Intergovernmental Panel on Climate Change (IPCC)*, 2001]. It has been suggested that their effects might be sufficient to moderate the positive forcing by anthropogenic greenhouse gases on regional scales [*Mitchell et al.*, 1995; *Krishnan and Ramanathan*, 2002]. The increased atmospheric sulfate in southern Asia is already implicated as a cause of local surface temperature declines and reductions in monsoon rainfall [*Krishnan and Ramanathan*, 2002; *Ramanathan et al.*, 2005; *Meehl et al.*, 1996]. Thus, knowledge of the distribution and concentration of sulfate aerosols is required to model and evaluate any associated climate changes [*Karoly et al.*, 2003; *Levitus et al.*, 2001]. Unlike the

long-lived greenhouse gases which are distributed uniformly over the globe, sulfate lifetimes are generally a week or less [*IPCC*, 2001; *Ramanathan et al.*, 2001], resulting in substantial spatial and temporal variations. Although sulfate histories have been reconstructed from polar ice cores [*Fischer et al.*, 1998; *Mayewski et al.*, 1990], no comparable records exist for tropical and subtropical regions and this limits effort to study the global sulfate cycle and its role in regional climate changes.

[3] Ice core records from Greenland and the European Alps suggest that atmospheric sulfate has increased since the Industrial Revolution, primarily due to increased SO_2 emissions from North America and Europe [*Fischer et al.*, 1998; *Mayewski et al.*, 1990]. However, since 1970 sulfate concentrations in the Greenland cores have decreased due to air pollution control measures in North America and Europe [*Fischer et al.*, 1998; *Mayewski et al.*, 1990]. In Asia, although the fossil energy-related SO_2 release is nearly an order of magnitude smaller than that in North America and Europe [*Smith et al.*, 2001], Asia is rapidly catching up. Roughly 40% of the world's population lives in southern and eastern Asia, and hence the potential for increased pollutant emissions is high and closely linked to the growing demand for energy. Although variations in sulfate emissions over Asia have been estimated for the last few decades [*Smith et al.*, 2001; *Sarath et al.*, 2003], little is known about the longer term trends. Here we present a 1000-year record of sulfate concentrations over southern Asia, deduced from a high resolution ice core drilled on the Dasuopu glacier in the central Himalayas.

2. Data and Results

[4] Dasuopu glacier in the central Himalayas is surrounded by highly populated and rapidly developing countries, such as India and China (Figure 1), it is an ideal location for extracting a history of regional anthropogenic emissions. Dasuopu is located downwind from southern Asia during the monsoon season and it is a sensitive recorder of fluctuations in the intensity of the south Asian monsoon as it receives up to 70% of its annual snowfall during that time [*Thompson et al.*, 2000; *Duan et al.*, 2004]. Observations show that the pollutants over the high Himalaya originate from distant sources in South Asia, some as far away as the India plains [*Edward and Bidur*, 2002; *Shrestha et al.*, 2000]. The summer monsoon circulation brings considerable pollution from South Asia to the Himalayas. During winter and the pre-monsoon period the large-scale circulation patterns are more dominated by westerly flow. These air masses do not bring large quantities of pollutants into the Himalaya as they are long traveled from urban and industrialized regions and have much lower concentrations of pollutants when compared to southerly

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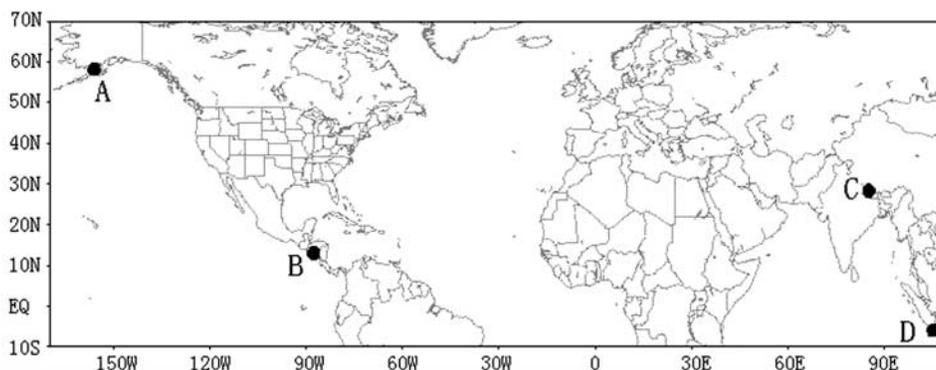
Figure 1. Location map of Novarupta volcano (A), Coseguina volcano (B), Dasuopu ice core site (C), and Krakatau volcano (D).

air masses [*Edward and Bidur*, 2002; *Shrestha et al.*, 2000]. During the winter and pre-monsoon season, a significant trans-Himalayan surface air flow from Nepal to Tibet occurs in the afternoon and evenings as a result of valley wind systems and large circulation patterns [*Edward and Bidur*, 2002; *Shrestha et al.*, 2000]. Due to the lack of precipitation (the air is dry at this time of year) there is a gradual accumulation of pollutants. During the pre- and early monsoon periods, pollutants are transported to higher elevations by convection where the upper level horizontal flow transports them northward over the Himalayas where they are deposited. Thus, the ice cores from the Himalayas provide a history of the ionic concentrations of these pollutants, including sulfate, from southern Asian.

[5] In 1997, three ice cores were collected from the Dasuopu glacier (28°23′N; 85°43′E; 7200 m asl) in the central Himalayas [*Thompson et al.*, 2000]. The results presented here are from Core 3, which was analyzed for major anion species (SO_4^{2-}, NO_3^- and Cl^-) and annually dated for the last 560 years and decadally dated for the last 1000 years [*Thompson et al.*, 2000]. Figure 2 shows the annual sulfate concentration for the most recent well-dated 396 years and decadal average for the last 1000 years. The annual time series exhibits variability on both the interannual and inter-decadal timescales. A number of spikes are apparent in the annually averaged sulfate concentrations and likely reflect either volcanic or drought events.

[6] A noticeable rise in the sulfate concentration baseline is observed in the middle of 19th century, with the largest rate of increase occurring after 1930. Annually averaged sulfate concentrations doubled from ~24 ppb in the first half of 19th century to ~49 ppb for the period from 1930 to 1997 and more than tripled to ~83 ppb for the period from 1990 to 1997. Concentrations episodically exceed 100 ppb. The decadal trends in annual sulfate concentration for different time intervals are compared in Table 1. From 1000 to 1870 the trend is near zero. However, the 1871 to 1997 trend of 3 ppb per decade is statistically significant at the 95% confidence level. This rate of increase has doubled to an average of 6 ppb per decade from 1930 to 1997, but this accelerated to 15 ppb per decade from 1970 to 1997 (both sig. at the 99% level).

[7] The most likely sources of sulfate in Himalayan precipitation are from sea spray, mineral dust, oxidation of sulfur emitted by the marine biota (DMS), volcanic emis-

sions via SO_2 activity and anthropogenic emissions via SO_2. Sodium and calcium concentrations, generally used to estimate the contribution of sea-salt and mineral dust to the observed sulfate concentrations [*Schwikowski et al.*, 1999; *Wang et al.*, 2002; *Kang et al.*, 2000], have remained constant over the last 200 years as confirmed by analysis of Dasuopu Core 2 (not shown). This indicates a constant sulfate contribution from sea-salt and mineral dust over the period. Since 1600 AD many of the large perturbations are associated with known volcanic eruptions such as Coseguina (1835), Krakatau (1883) and Novarupta (1912) (Figure 2). Sulfate peaks may also result from spring/summer dust storms [*Kang et al.*, 2000; *Gao et al.*, 1992]
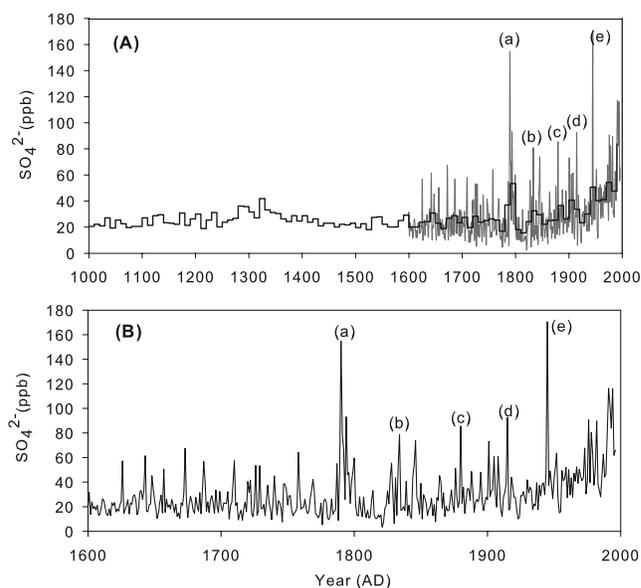

Figure 2. (a) Annual and decadal average sulfate concentration (ppb) for the Dasuopu ice core, central Himalayas, during the period 1600–1997 and 1000–1997, respectively. (b) Annual sulfate concentration (ppb) from 1600 to present in the enlargement. Examples of volcanic events recorded by sulfate spikes are: Coseguina (1835, spike b); Krakatau (1883, spike c); Novarupta (1912, spike d). Spike a reflects a monsoon failure and the source for spike e is unknown.

Table 1. Trends in Sulfate Concentration at the Dasuopu Ice Core Site

Time Interval	1000–1870	1871–1996	1930–1996	1970–1997
Trend, ppb per decade	0.002	3	6	15
Confidence level, %	50	95	95	99

and complicate interpretation of the volcanic history. For example, the sulfate spikes in the 1330s and from 1790 to 1796 reflect known Indian monsoon failures [*Thompson et al.*, 2000]. Nevertheless, the sporadic sulfate spikes do not greatly alter either the mean concentrations or the trends. Lack of long-term records of natural sulfate contributions necessitates assuming that the natural contributors to sulfate today are comparable to those prior to 1870. Thus, the main sources of the increased sulfate concentrations since 1870s in the Dasuopu ice core are almost certainly anthropogenic in origin.

[8] Organic compounds and lead, extracted from snow pits and shallow ice cores on the Dasuopu glacier in 1997, reveal that this remote Himalayan region and the middle to upper troposphere are polluted by the byproducts of human activities [*Ramanathan et al.*, 2005; *Huo et al.*, 1999; *Xie et al.*, 2000], primarily from the increased burning of fossil fuels in India. The 1000-year sulfate record from the Dasuopu ice core provides not only the potential for a history of human pollution in the Himalayan region, but also the history of pre-industrial, natural sulfate levels which were generally very low and constant in this remote region. The sulfate concentration (Figure 2) shows little change from 1000 to 1870 A.D. but as discussed above has increased by a factor of three since the 1870s. Since the 1930s anthropogenic sulfate emissions appear to have dominated the natural sulfate cycle in the region and now affect both the local climate and environment [*Krishnan and Ramanathan*, 2002; *Ramanathan et al.*, 2005; *Meehl et al.*, 1996].

3. Discussion and Conclusion

[9] The Dasuopu ice core presented here represents the lowest latitude, ice-core-derived sulfate record recovered in the Northern Hemisphere and when compared with the high latitude sulfate history extracted from Greenland ice cores, interesting temporal and spatial patterns of atmospheric sulfate concentrations emerge. Figure 3 shows the long trends detected by singular spectrum analysis (SSA) in both the Dasuopu ice core and the B16 Greenland core [*Fischer et al.*, 1998]. The common features of their sulfate records are: (1) the lack of obvious trends before industrialization, (2) an increasing trend starting around 1870–1880 AD, (3) a transient drop during the 1930s, and (4) a major increase in sulfate background levels starting around the 1940s (Figure 3). Their similar trends reflect the large-scale anthropogenic release of sulfate over Europe, North America and southern Asia since the Industrial Revolution. Sulfate concentrations in the Dasuopu ice core, however, are much lower than those in the Greenland B16 ice core [*Fischer et al.*, 1998] for both the pre-industrial and industrial periods. The sulfate concentration in the Greenland core doubled from 1870 to 1920, while that in Dasuopu increased only 40% over the same period. After 1970, the two ice core records show dramatic differences. The sulfate concentration in the Greenland ice core peaked during the late 1970s and dropped by nearly 30% thereafter [*Fischer et al.*, 1998], likely reflecting strict environmental regulation in the United States and Europe. In contrast, the sulfate concentrations in the Dasuopu core nearly doubled after 1970. This post-1970 difference likely reflects regional differences in source strength and transport pathways for atmospheric sulfate between Europe and Asia, as well as regional differences in the degree of environmental regulation.

[10] Comparisons of the Greenland sulfate records with SO_2 emission estimates for the United States and Europe suggest that Greenland sulfate levels were controlled mainly by North American emissions in the first half of the twentieth century, whereas the major increase detected after 1950 originates mainly from European emissions [*Fischer et al.*, 1998; *Mayewski et al.*, 1990]. The difference in sulfate deposition trends between Dasuopu and Greenland since 1970 almost certainly reflects differing regional source strengths. To examine this, the Dasuopu and Greenland sulfate trends, detected using SSA, are compared with SO_2 emissions estimated from energy and industry statistics for Europe, the United States and southern Asia [*Smith et al.*, 2001; *Gschwandtner et al.*, 1986; *Mylona*, 1996] (Figure 3). The 1970 peak in Greenland sulfate concentrations is also evident in the SO_2 emissions from Europe, (and to a much lesser extent also in those from the U.S.), pointing to European anthropogenic sources as the primary contributors to sulfate levels in Greenland ice since the 1950s. The differing trends for sulfate deposition on Dasuopu and for SO_2 emissions in Europe and the United States clearly point to another important SO_2 source. The close correspondence of the sulfate trend on Dasuopu and SO_2 emissions over southern Asia (Figure 3a) indicate that south Asian anthropogenic sources have dominated the concentrations of sulfate levels deposited on Himalayan
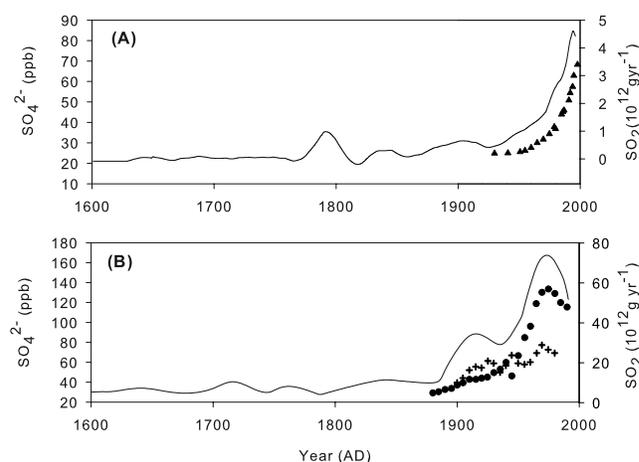
Figure 3. (a) Sulfate trend concentrations (ppb) in the Dasuopu ice core and SO_2 emissions (triangle) (10^{12} g yr^{-1} or megatons) over southern Asia [*Smith et al.*, 2001]. (b) Sulfate trend concentrations (ppb) in the Greenland B16 ice core [*Fischer et al.*, 1998] and the anthropogenic SO2 emissions for the United States (cross) and Eurasia (dot) [*Gschwandtner et al.*, 1986; *Mylona*, 1996].

glaciers since the 1930s. Figure 3 reveals that sulfate concentrations in the Dasuopu ice core increased by a factor of 2.5 from 1951–1997 while SO_2 emissions over southern Asia increased 6 fold over the same period [*Smith et al.*, 2001].

[11] Although SO_2 emissions in southern Asian are small relative to the total global output [*Smith et al.*, 2001], anticipated economic development in that region, coupled with the lack of emission regulations, indicate the likelihood of a continued acceleration of SO_2 emissions and hence regional pollution. The snow accumulating annually on Himalayan ice fields, such as the Dasupou glacier, provides an excellent recording system that can monitor current anthropogenic emissions and place their current concentration levels within a longer-term, pre-anthropogenic, natural context.

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