

## Annually resolved southern hemisphere volcanic history from two Antarctic ice cores

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**Abstract.** The continuous sulfate analysis of two Antarctic ice cores, one from the Antarctic Peninsula region and one from West Antarctica, provides an annually resolved proxy history of southern hemisphere volcanism since early in the 15th century. The dating is accurate within  $\pm 3$  years due to the high rate of snow accumulation at both core sites and the small sample sizes used for analysis. The two sulfate records are consistent with each other. A systematic and objective method of separating outstanding sulfate events from the background sulfate flux is proposed and used to identify all volcanic signals. The resulting volcanic chronology covering 1417–1989 A.D. resolves temporal ambiguities about several recently discovered events. A number of previously unknown, moderate eruptions during late 1600s are uncovered in this chronology. The eruption of Tambora (1815) and the recently discovered eruption of Kuwae (1453) in the tropical South Pacific injected the greatest amount of sulfur dioxide into the southern hemisphere stratosphere during the last half millennium. A technique for comparing the magnitude of volcanic events preserved within different ice cores is developed using normalized sulfate flux. For the same eruptions the variability of the volcanic sulfate flux between the cores is within  $\pm 20\%$  of the sulfate flux from the Tambora eruption.

### Introduction

Explosive volcanic eruptions introduce large amounts of dust and gaseous materials into the atmosphere and often directly into the stratosphere. The main component of acidic volcanic gases, sulfur dioxide ( $\text{SO}_2$ ), is oxidized in the atmosphere to form sulfuric acid and water ( $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ ) aerosol particles. In the stratosphere these aerosol particles are spread globally via atmospheric circulation and often reside there for several years during which they may alter significantly the radiative balance and albedo of the Earth-atmosphere system. Satellite and ground-based measurements of the June 1991 eruption of the Mount Pinatubo volcano ( $15^\circ\text{N}$ ,  $121^\circ\text{E}$ ) in the Philippines clearly show a rapid zonal dispersion of sulfurous aerosols [Bluth *et al.*, 1992] and then a gradual poleward transport [Trepte *et al.*, 1993].

Explosive volcanism has long been recognized as one of the three most important factors affecting the decadal to millennial scale climate variability (the other two are solar variability and internal interactions of the ocean-atmosphere system). The short-term climatic impact of explosive volcanic eruptions has been well documented [Rampino and

Self, 1982; Robock, 1991]. For example, following the Pinatubo eruption, a decrease of  $0.2\text{--}0.7^\circ\text{C}$  was observed in global tropospheric and near-surface temperatures [McCormick *et al.*, 1995; Robock and Mao, 1995] with a corresponding increase in stratospheric temperatures [Randel *et al.*, 1995]. In general, the temperature responses to explosive eruptions last several years [Robock, 1991; Robock and Mao, 1995].

Evaluating the climatic (e.g., temperature) effects of volcanism requires comprehensive and accurate climatic and volcanic histories. While historical temperature records are fairly extensive [e.g., Jones, 1994], the history of global volcanism is, in most cases, sparse and only qualitative or semiquantitative. Existing volcanic chronologies include the dust veil index (DVI) of Lamb [1970] and the volcanic explosivity index (VEI) by Newall and Self [1982] and Simkin *et al.* [1981]. These indices rely on historical and geological compilations and therefore suffer from incomplete early records and historical inaccuracies [Robock and Free, 1995]. An ideal volcanic history for the purpose of evaluating the volcanic impact on the climate should (1) provide a quantitative proxy of the atmospheric effects caused by volcanic eruptions, (2) include a comprehensive and accurate chronology of the most explosive eruptions, and (3) contain high temporal resolution for accurate dating of the eruptions.

Recently polar ice cores, along with tree-ring data [e.g., Jones *et al.*, 1995, and references therein], have provided most of the newly acquired information on volcanic history. The concentrations of sulfuric acid ( $\text{H}_2\text{SO}_4$ ), sulfate ( $\text{SO}_4^{2-}$ ), and acidic conductivity of Antarctic ice cores have been shown to be directly related to aerosol concentrations of  $\text{H}_2\text{SO}_4$  in the atmosphere [e.g., Wagenbach *et al.*, 1988]. Large amounts of  $\text{SO}_2$  from volcanic eruptions enhance

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atmospheric sulfur concentrations from months to a few years. Hammer [1977] and Hammer *et al.* [1980] first used the electrical conductivity method (ECM) to measure the acidity of Greenland ice cores as a proxy for volcanic history. Legrand and Delmas [1987] proposed a glaciological volcanic index (GVI) based on  $\text{H}_2\text{SO}_4$  concentrations in ice cores as a way to estimate the global impact of explosive eruptions. Robock and Free [1995] evaluated the strengths and limitations of combining various ice core records to create an ice core-volcano index (IVI). Zielinski *et al.* [1994] recently published  $\text{SO}_4^{2-}$  measurements from the central Greenland GISP2 ice core, which have provided the longest continuous (biyearly resolution) volcanic record from an ice core.

For the southern hemisphere (SH), Delmas *et al.* [1992] constructed a 1000-year volcanic record based on ECM and  $\text{H}_2\text{SO}_4$  measurements on a number of south pole ice cores. Moore *et al.* [1991] measured continuously the dielectrical conductivity of an ice core from East Dronning Maud Land (site G15) in East Antarctica and presented the results as a 770-year volcanic record. Langway *et al.* [1994, 1995] reported a 1300-year record from a recent core from Byrd Station in West Antarctica.

All of the volcanic horizons in these previous Antarctic ice cores were detected by an initial survey of the cores using ECM or the dielectric profiling (DEP) method. Ice layers containing suspected "volcanic events" were subsequently analyzed for  $\text{H}_2\text{SO}_4$  or  $\text{SO}_4^{2-}$  concentrations. Although major eruptions are detected in a quick scan of ice cores using the ECM and DEP methods, small and moderate volcanic signals may be overlooked due to the background variability of the signals which these methods measure. The ECM method detects the ice acidity which may have significant contributions from acids other than  $\text{H}_2\text{SO}_4$  (e.g.,  $\text{HNO}_3$  and carboxylic acids). The DEP method is an indirect measurement of total salt content of the ice [Moore *et al.*, 1992]. Nonvolcanic ice layers containing unusually large amounts of snow impurities other than  $\text{H}_2\text{SO}_4$  and  $\text{SO}_4^{2-}$  may exhibit high ECM and DEP signals and may thus be erroneously identified as volcanic. Furthermore, most of these volcanic chronologies rely on mathematical modeling to establish the depth-age relationship. In general, the modeling approach assumes a constant snow accumulation rate and establishes the depth-age relationship based on the physics of ice flow and densification. This relationship may be refined further using known volcanic time-stratigraphic horizons, such as Tambora (1815) and the 1259 unknown eruption first identified by Langway *et al.* [1988]. As accumulation rates may vary substantially over a few decades or longer and identification of the exact eruption producing the volcanic  $\text{SO}_4^{2-}$  signal is difficult, the timescales based on this method may contain considerable errors and uncertainties.

We present  $\text{SO}_4^{2-}$  records from two ice cores: one from Siple Station ( $75^\circ 55'S$ ;  $84^\circ 15'W$ ) located at the base of the Antarctic Peninsula, the other from the Dyer Plateau ( $70^\circ 40'S$ ;  $64^\circ 52'W$ ), about halfway between the northern tip and the southern base of the peninsula (Figure 1). These two  $\text{SO}_4^{2-}$  records are used to construct an annually resolved, continuous volcanic chronology for the SH. This chronology offers improvements upon some of the previously published ice core-derived volcanic records: (1) the parameter or proxy

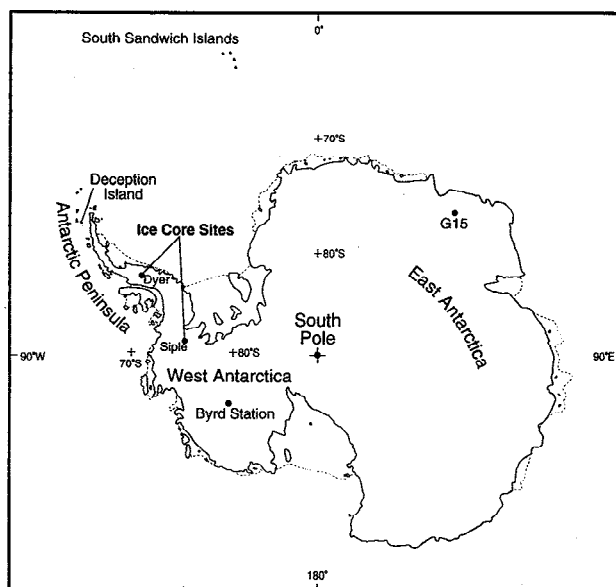


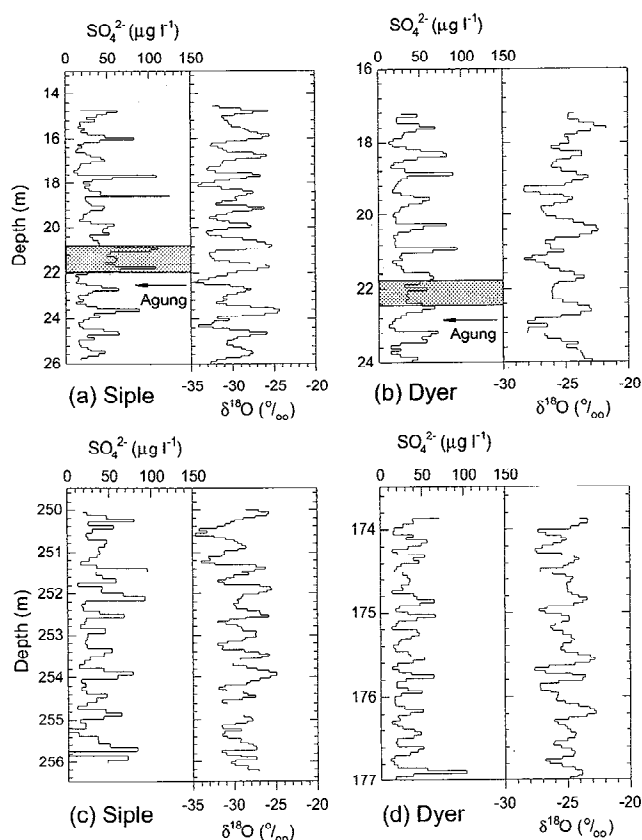
Figure 1. Location of Siple Station and the Dyer Plateau in the Antarctic Peninsula region and other Antarctic ice core sites.

for volcanic signals is the quantity of  $\text{SO}_4^{2-}$ , which is related directly to  $\text{SO}_2$ , the major chemical component emitted by volcanic eruptions, whereas the acidity in Antarctic snow/ice may be affected by the presence of nonvolcanic acids; (2) both records are dated entirely by layer counting, so the dating accuracy of identified volcanic events is significantly higher than in the previous records based on modeled depth-age relationships; and (3) the ice core records from two different sites are combined which helps eliminate spurious signals arising from postdepositional effects, poor ice core quality, and other experimental errors. In addition, these records are from the West Antarctica and Antarctic Peninsula region where few long, high-resolution volcanic records are currently available.

### Analysis and Dating of Cores

The Siple core (302 m) was drilled in the 1985-1986 austral summer, while the two 235-m Dyer cores (cores 1 and 2) were drilled, 1 meter apart, in 1989-1990. The Siple core was analyzed continuously for concentrations of major anions ( $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ ), oxygen isotopic ratios ( $\delta^{18}\text{O}$ ) and microparticle concentrations. The top 181 m (0-112 m of core 1 and 107-181 m of core 2) of the Dyer cores were similarly sampled and analyzed. Procedures of sample preparation and ion chromatographic measurements are presented elsewhere [Dai *et al.*, 1995]. Concentrations of major cations ( $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ ) and aluminum (Al) were measured on selected core sections using a Varian SpectrAA-20 atomic absorption spectrophotometer with a GTA-96 graphite furnace atomizer (GFAA).

To resolve an annual layer, five to eight measurements are required for a seasonally varying parameter (e.g.,  $\delta^{18}\text{O}$  or  $\text{SO}_4^{2-}$ ). Because of the relatively high annual snow accumulation at Siple (~550 mm of water equivalent (w.e.)) and Dyer (~450 mm w.e.), it was possible to cut an average of



**Figure 2.** Annual (seasonal) variations of  $\text{SO}_4^{2-}$  concentrations in microgram per liter ( $\mu\text{g L}^{-1}$ ) and  $\delta^{18}\text{O}$  in parts per thousand (‰) are shown at the top of the Siple (a) and the Dyer (b) cores and near the bottom of the Siple (c) and the Dyer (d) cores. The arrows in Figures 2a and 2b point to the probable time of the Agung eruption (1963), and the shaded area indicates the time period when volcanic  $\text{SO}_4^{2-}$  deposition from Agung eruption likely occurred.

seven to eight samples (approximately fifteen samples at the top and five samples at the bottom) for each annual layer in both cores. This high sampling frequency (resolution) easily resolved the well-preserved  $\delta^{18}\text{O}$  and  $\text{SO}_4^{2-}$  seasonal cycles (Figure 2) which were counted to establish the depth-age relationship with excellent accuracy (e.g., the 1815 Tambora eruption was dated both in the Siple and in the Dyer cores at 1816 and 1817, as expected). The accumulated errors, attributable to a few ambiguous seasonal cycles, are estimated to be only  $\pm 3$  years at the end of each record. The 302-m Siple core contains 568 years (1417–1985 A.D.), while the Dyer cores yield a 485-year (1504 to 1989) continuous chronology [Dai *et al.*, 1995]. Here all dates relating to ice cores are considered "ice core years," such that 1988 represents the snow layer accumulated from the austral winter (most negative  $\delta^{18}\text{O}$  during that seasonal cycle) of 1987 to the austral winter of 1988.

## Data Presentation

### Non-Sea-Salt $\text{SO}_4^{2-}$

The extremely low concentrations of Al and microparticles in both Siple and Dyer samples indicate that there is little continental aerosol dust in the snow [Mosley-Thompson *et al.*, 1991; Thompson *et al.*, 1994]. Therefore sea-salt

aerosols are assumed to be the only important source of  $\text{Na}^+$  in the snow. The ratios of  $\text{Na}^+$  to  $\text{Cl}^-$  concentrations in Siple and Dyer snow are similar to that of bulk seawater [Dai *et al.*, 1995], suggesting that like  $\text{Na}^+$ , essentially all  $\text{Cl}^-$  is also derived from sea-salt aerosols. Therefore non-sea-salt (nss)  $\text{SO}_4^{2-}$  concentrations are calculated using the  $\text{SO}_4^{2-}/\text{Cl}^-$  concentration ratio in bulk sea water. Because nss  $\text{SO}_4^{2-}$  constitutes the overwhelming majority of total  $\text{SO}_4^{2-}$  ( $\geq 95\%$ ) at these two locations, no distinction is made between total  $\text{SO}_4^{2-}$  and nss  $\text{SO}_4^{2-}$  in subsequent discussion, although the volcanic  $\text{SO}_4^{2-}$  flux calculations are all based on nss  $\text{SO}_4^{2-}$ .

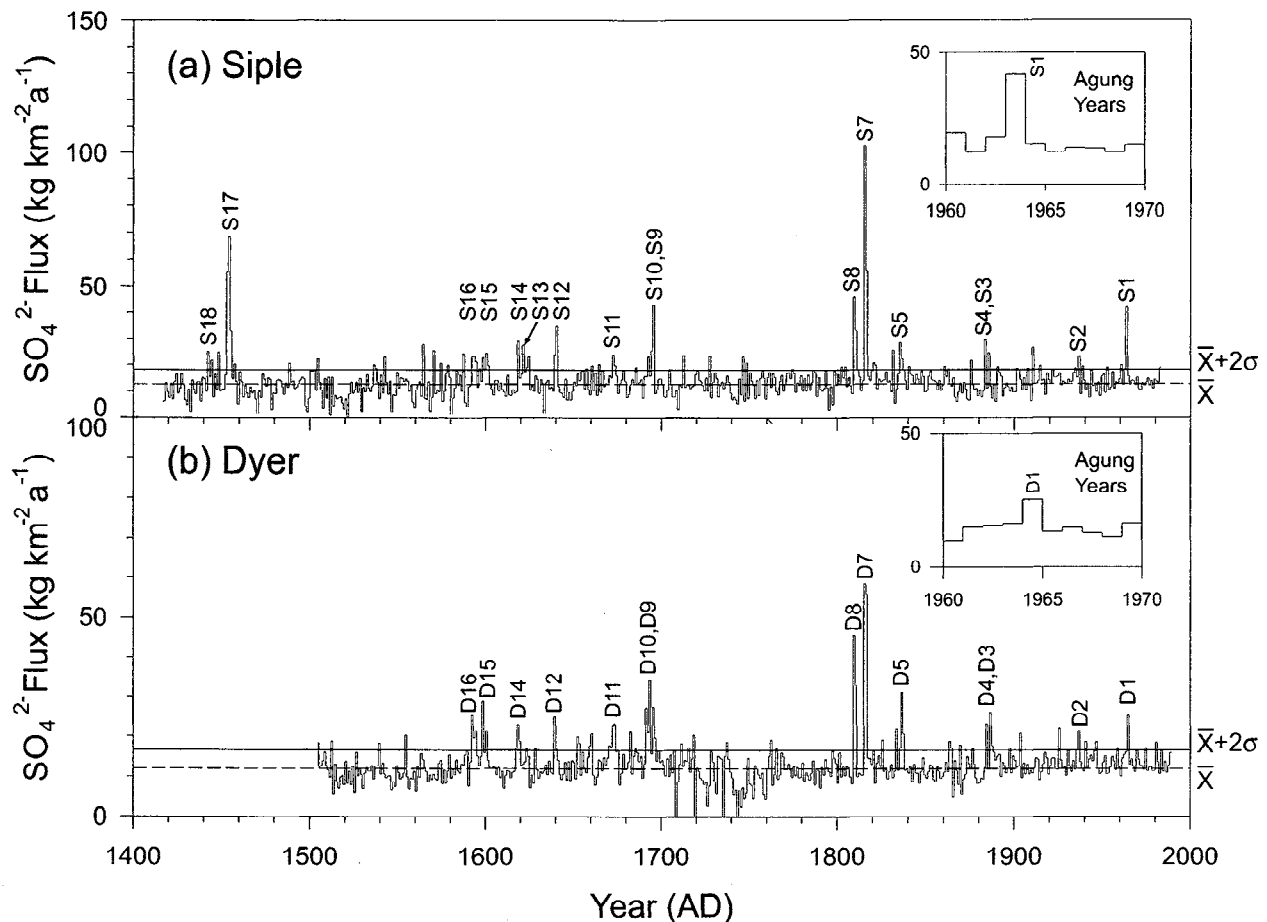
### Calculation of Annual $\text{SO}_4^{2-}$ Flux

With a subannual sampling frequency, it was possible to calculate the annual  $\text{SO}_4^{2-}$  flux for the Dyer cores [Dai *et al.*, 1995] and the Siple core. Because of ice flow and densification, annual layers become progressively thinner with depth. Because flux calculations depend on sample or layer thickness, it is necessary to adjust the measured layer thicknesses to account for layer thinning. In the Dyer cores the original annual layer thicknesses were reconstructed by ice flow modeling using ice depth and strain rates measured in the field [Weertman, 1993]. No ice flow and strain rate measurements are available for Siple Station and the depth of the ice sheet can only be estimated from airborne radio echo soundings by Scott Polar Research Institute [see Drewry, 1983, Figure 2c]. Using a simple flow model and assuming a constant accumulation rate (550 mm w.e.) and an ice depth of 1800 m, Mosley-Thompson *et al.* [1993] reconstructed the original thicknesses of individual annual layers in the Siple core. The annual  $\text{SO}_4^{2-}$  fluxes in the Siple and Dyer cores were subsequently calculated using these reconstructed annual layer thicknesses. The unsmoothed data, shown in Figure 3, form the basis of the ice-core-derived volcanic chronology presented below.

No continuous ice core  $\text{SO}_4^{2-}$  flux records have been reported previously, although several recent ice core studies [Delmas *et al.*, 1992; Zielinski *et al.*, 1994] have calculated the  $\text{H}_2\text{SO}_4$  or  $\text{SO}_4^{2-}$  fluxes for specific volcanic events. Our results presented below indicate that using the annual  $\text{SO}_4^{2-}$  flux and its background variability facilitates the detection of small and moderate volcanic signals. Therefore our data are presented as annual  $\text{SO}_4^{2-}$  fluxes rather than concentrations. However, as the relationship between the concentrations and fluxes of snow impurities and their abundance in the atmospheric reservoir remains poorly understood at this time, it is unclear whether concentrations or fluxes provide a better proxy for the atmospheric abundance.

## Results and Discussion

Volcanic aerosols are transported and distributed by the atmosphere prior to deposition in snow. Atmospheric phenomena such as the Quasi-Biennial Oscillation can strongly influence the direction and speed of the global spread of aerosols from low-latitude eruptions, as occurred with Pinatubo [Trepte *et al.*, 1993]. Therefore the temporal variation of volcanic signals in ice cores may reflect variations in the atmospheric circulation. However, the role of the atmospheric processes in the transport of volcanic aerosols to the polar ice sheets is beyond the scope of this study, which focuses on the glaciological variability of volcanic signals.



**Figure 3.** Annual nss  $\text{SO}_4^{2-}$  fluxes are shown for the Siple (a) and Dyer (b) cores. Data are unsmoothed. Dashed straight lines represent average nonvolcanic flux and the solid lines represent the upper limit of nonvolvanic flux ( $\bar{x}+2\sigma$ ). The inserts show years in which the Agung eruption appears. A section in the Dyer core (112–130 m) suffered loss of ice during core drilling, resulting in low fluxes from 1706 to 1763 A.D. in Figure 3b. See Dai *et al.* [1995] for details.

### Background Versus Volcanic $\text{SO}_4^{2-}$

Organic sulfur compounds (e.g., dimethyl sulfide or DMS) from marine biogenic emissions permeate the SH marine atmosphere. The final oxidation product of these compounds is primarily  $\text{SO}_4^{2-}$  [Saltzman, 1995], which forms the background  $\text{SO}_4^{2-}$  in Antarctic snowfall. Volcanic  $\text{SO}_4^{2-}$  is superimposed on this variable background and a useful ice core volcanic chronology must separate volcanic  $\text{SO}_4^{2-}$  from the background  $\text{SO}_4^{2-}$  in the snow.

The detection of volcanic signals in ice cores, especially those corresponding to the small and moderate events, depends on how the nonvolcanic background is estimated. Crowley *et al.* [1993] used the signal from the 1883 Krakatoa eruption as a minimum reference to exclude the varying background in the acidity record from the Crête (Greenland) ice core by Hammer *et al.* [1980]. A number of quantitative strategies have also been used to separate volcanic signals from the background. Delmas *et al.* [1992] defined a volcanic signal as a  $\text{SO}_4^{2-}$  concentration spike larger than the “normal” background (average plus standard deviation). Zielinski *et al.* [1994] approximated the background  $\text{SO}_4^{2-}$  concentrations in the GISP2 core using a spline-fitted curve and then considered events to be volcanic when  $\text{SO}_4^{2-}$

concentrations exceed the value of the fitted background plus a standard deviation of the residuals.

The “normal” background approach of estimating nonvolcanic  $\text{SO}_4^{2-}$  and its variability is based on the assumption that in the absence of significant inputs from volcanic and anthropogenic  $\text{SO}_4^{2-}$  the  $\text{SO}_4^{2-}$  flux and concentration in snow are not controlled by any dominant sources or transport and deposition processes. Legrand and Feniet-Saigne [1991] found a significant correlation between the concentrations of methanesulfonic acid (MSA) in a south pole ice core and the El Niño Southern Oscillation (ENSO) events. As MSA and nonvolcanic  $\text{SO}_4^{2-}$  share the same source (DMS), it was suggested that nonvolcanic  $\text{SO}_4^{2-}$  in Antarctic snow may be related to ENSO variations. However, in the same south pole core, no similar relationship was found between  $\text{SO}_4^{2-}$  concentration and ENSO, indicating that background  $\text{SO}_4^{2-}$  is not affected by the phrases of ENSO, a coupled atmosphere-ocean phenomenon that may influence the transport of ocean-emitted sulfur species to the Antarctic troposphere. Pasteur *et al.* [1995] reported no significant link between either MSA or nss  $\text{SO}_4^{2-}$  and ENSO events in an ice core from Dolleman Island on the Weddell Sea side of the Antarctic Peninsula. The background  $\text{SO}_4^{2-}$  at Siple and Dyer, which are in the

same region as Dolleman Island, is likely to be similar to that of Dolleman Island in terms of potential oceanic influence, and therefore is not likely affected by ENSO events.

**Flux Versus Concentration as a Tool to Facilitate the Detection of Volcanic Signals**

To assess the nonvolcanic annual SO<sub>4</sub><sup>2-</sup> flux background and the range of its variations, the following procedure is devised. The annual flux data are first smoothed with a three-sample running mean to remove noise originating from the division of samples into annual layers. Next, those years associated with either a known or probable large volcanic eruption (marked with an asterisk in Table 1) are removed from the annual flux time series. The identification of these volcanic events relied on the accurate dating of the Siple and Dyer cores and was made using the VEI list of volcanic records [Newall and Self, 1982] and with reference to results from previous Antarctic ice cores. Finally, the average ( $\bar{x}$ ) and standard deviation ( $\sigma$ ) of the remaining smoothed annual fluxes are computed to represent the background and its variability, respectively. Subsequently, volcanic years are identified as those having a SO<sub>4</sub><sup>2-</sup> flux that exceeds the background ( $\bar{x}$ ) by 2 $\sigma$ , an approach similar to that used by Delmas et al. [1992]. Fortunately, the anthropogenic SO<sub>4</sub><sup>2-</sup> influence in the SH is minimal, making it unnecessary to detrend the Siple and Dyer data for the last 100 years, as is necessary for Greenland ice cores [Robock and Free, 1995].

An alternative to the flux procedure is using the SO<sub>4</sub><sup>2-</sup> concentration data to detect volcanic signals. However, the variability of the background SO<sub>4</sub><sup>2-</sup> concentrations at Dyer

and Siple is considerably higher than that for the SO<sub>4</sub><sup>2-</sup> annual fluxes: the relative standard deviation (RSD=%(standard deviation/mean), which allows comparison of variability between different data sets) is  $\approx$ 70% for the overall nonvolcanic SO<sub>4</sub><sup>2-</sup> concentrations in both the Dyer and the Siple cores, compared with  $\approx$ 30% for the annual fluxes. This higher variability of sample concentrations makes it more difficult to detect small and moderate volcanic events, even though a similar approach used by Delmas et al. [1992] appears to have been effective for south pole ice cores. To compare this concentration approach with the flux procedure described above, the average of the annual nonvolcanic SO<sub>4</sub><sup>2-</sup> maxima and its standard deviation in each of the Dyer and Siple data sets were used to identify potential volcanic signals (concentrations are considered volcanic only when they exceed the nonvolcanic annual maxima). Only the largest eruptions (Tambora, the 1809 Unknown, Huaynaputina, and Kuwae in Table 1) were identified. In contrast, the flux procedure produced a more extensive list of volcanic events (Table 1), including all those found in the south pole cores by Delmas et al. [1992] and four previously unreported events (see discussion on individual events).

The effect of the flux approach is further illustrated by the identification of the Agung eruption (1963). As shown in Figures 2a and 2b, the summer SO<sub>4</sub><sup>2-</sup> concentrations associated with the 1963 Agung eruption (shaded) do not exceed the normal annual summer SO<sub>4</sub><sup>2-</sup> maxima. Only the winter SO<sub>4</sub><sup>2-</sup> concentrations, which are lower than the summer maxima, in 1964-1965 are above the average winter concentrations for nonvolcanic years. Thus the Agung signal could not be identified using only high SO<sub>4</sub><sup>2-</sup> concentrations. As illustrated in the inserts in Figure 3, the Agung signal is easily recognizable in the flux profiles. For Dyer and Siple, small and moderate volcanic signals can be detected only by a close examination of the detailed seasonal SO<sub>4</sub><sup>2-</sup> concentrations or the calculation of annual flux. In both cases a subannual sampling frequency is required.

**Identification of Volcanic Years and Events**

The flux procedure described above was used to produce a list of potential volcanic years from the Siple and Dyer SO<sub>4</sub><sup>2-</sup> flux data sets. Since smoothing "spreads" an original volcanic year into the adjacent years, the smoothed data sets were examined against the unsmoothed data sets to remove false-positive years from the list. The remaining years were considered volcanic in origin. Consecutive volcanic years may result from either a large eruption producing multiple-year SO<sub>4</sub><sup>2-</sup> deposition or two or more eruptions occurring within a short (1 or 2 years) time interval. It is probable that a careful examination of the SO<sub>4</sub><sup>2-</sup> concentrations of the individual samples in the volcanic years may be the only reliable way to distinguish between these two scenarios. Fortunately, the high sampling frequency and dating accuracy of the Siple and Dyer cores allow the resolution of closely spaced events that were difficult to resolve in previous ice core records [Delmas et al., 1992; Langway et al., 1995; Legrand and Delmas, 1987; Moore et al., 1991].

Table 1 lists all potential volcanic events, selected using the procedure described above, for the past 570 years as recorded in these two Antarctic ice cores. In Table 1 and Figure 3 the events are numbered and denoted by "S" for

**Table 1. Volcanic Events Found in Siple and Dyer Ice Cores**

Siple Event	Event Date	Dyer Event	Event Date	Probable Eruption and Year
S1	1964-1965	D1	1965	*Agung 1963
S2	1937	D2	1937	? (Darney Island 1936)
S3	1886	D3	1887	*Tarawera 1886
S4	1884	D4	1885	*Krakatoa 1883
S5	1836-1837	D5	1836-1838	*Cosiguina 1835
S6	1832	D6	1834	
S7	1816-1817	D7	1816-1817	*Tambora 1815
S8	1810-1811	D8	1810-1811	*Unknown 1809
S9	1695-1696	D9	1696-1697	? (East Indies 1693-1694)
S10	1693	D10	1692	? (Reventador 1691)
S11	1673-1674	D11	1673-1674	? (San Salvador 1671 or Gamkonora 1673)
S12	1640-1641	D12	1640-1641	Awu 1640 and others
S13	1622	--	--	
S14	1619	D14	1619-1620	? sub-Antarctic 1618
S15	1599-1602	D15	1599-1601	Huaynaputina 1600
S16	1593-1595	D16	1593-1595	? (Raung 1592)
S17	1454-1457			*Kuwae 1453
S18	1443			?

See text for selection method. Event dates are ice core years in which volcanic SO<sub>4</sub><sup>2-</sup> deposit is found. Volcanoes are identified according to VEI and previously published records and unknown eruptions are represented by a question mark with the suspected eruptions in parentheses. Eruptions marked with an asterisk were removed from flux data sets to calculate nonvolcanic background. Dashes indicate that no contemporaneous event is found at Dyer.

Siple and "D" for Dyer. The event dates are years when the volcanic signals are found, not necessarily the eruption years. It may take 1 or 2 years for stratospheric  $\text{SO}_4^{2-}$  to be transported from lower latitudes to Antarctica. This lag is well documented for the Tambora eruption and is also evident for the June 1991 Pinatubo eruption (the Philippines), which first appears in south pole snow in late 1992 (J. Cole-Dai and E. Mosley-Thompson, unpublished data, 1995). A short lag (i.e., less than 1 year) indicates either a middle or high southern latitude location, while a long lag may indicate either a low-latitude location and hence a long transport time or a middle- to high-latitude eruption late in the austral summer or fall so that the volcanic aerosols will not penetrate the Antarctica troposphere until the breakup of the polar vortex the next spring. The individual events, along with the years of their appearance in the cores, are discussed in the following section. Contemporaneous events (within the range of dating error and uncertainty due to division of discrete years) in Siple and Dyer are presumed to have originated from the same eruption and therefore designated the same number.

Previously published records seem to indicate that the volcanic acid or  $\text{SO}_4^{2-}$  signals are more prominent in ice cores recovered from low snow accumulation areas. For example, the above mentioned Agung eruption appears prominently in Dome C and south pole cores [Legrand and Delmas, 1987], both located in the East Antarctica region where annual accumulation ( $\leq 100$  mm w.e.) is considerably lower than at Siple and Dyer. Sulfate deposition consists of wet (snow and fog) and dry deposition [Davidson et al., 1996]. In areas of relatively high snow accumulation the  $\text{SO}_4^{2-}$  contribution by dry deposition is small [Dai et al., 1995] and its concentration is probably diluted by high precipitation. Such a dilution effect may explain the reduced prominence of the Agung and other signals in cores from higher accumulation areas. However, the relationship between accumulation rate and the proportion of dry deposition is poorly understood and needs to be addressed in future research.

### Individual Events

**S1 (1964-65) and D1 (1965).** This event lasts nearly 2 years in both cores and is consistent with the 1963 Agung eruption ( $8^\circ\text{S}$ ,  $115^\circ\text{E}$ ). Since this eruption has been identified in many previous Antarctic ice cores [Delmas et al., 1985, and 1992; Langway et al., 1995; Legrand and Delmas, 1987; Moore et al., 1991], its signal may serve as a time-stratigraphic marker for future cores, although flux calculation may be necessary for its identification, as demonstrated previously (Figures 2 and 3). Despite the fact that Agung is located in the tropics and its  $\text{SO}_4^{2-}$  aerosols were expected to spread to both hemispheres, it has not been unambiguously identified in Greenland snow [Mosley-Thompson et al., 1993; Zielinski et al., 1994]. Two possible explanations are uneven interhemispheric distribution of the aerosols with the bulk going to the SH or that the  $\text{SO}_4^{2-}$  signal in Greenland is embedded in the high background concentrations due to the deposition of the NH anthropogenic  $\text{SO}_4^{2-}$ . Moderate NH eruptions in the same time period may also have obscured the Agung signal in Greenland [Zielinski et al., 1994]

**S2 (1937) and D2 (1937).** These are small signals (about 30% that of Agung and 8-10% of Tambora) and appear to

represent a single-year event. The VEI does not contain a significant eruption in the Southern Hemisphere in this time period, although the signal could have been left by a modest Antarctic or sub-Antarctic eruption, such as the VEI 2 eruption on Darney Island in the South Sandwich Islands ( $59^\circ\text{S}$ ,  $26^\circ\text{W}$ ) late in 1936, as recorded by Simkin et al. [1981]. At Siple this is a "one-sample" event (i.e., due to only one sample with high  $\text{SO}_4^{2-}$  concentration), while at Dyer, the  $\text{SO}_4^{2-}$  background is slightly elevated over a large part of the annual layer. This difference between Siple and Dyer may reflect the effect of postdepositional processes (e.g., snow drifting). If this signal is indeed produced by the Darney Island eruption, there may be another explanation for the difference: the strong and frequent storms at Dyer can prolong the  $\text{SO}_4^{2-}$  deposition process, whereas Siple lies more within the polar vortex, so the influx of the tropospheric volcanic  $\text{SO}_4^{2-}$  aerosols may be more sporadic.

**S3 (1886), D3 (1887) and S4 (1884), D4 (1885).** The elevated  $\text{SO}_4^{2-}$  flux and concentrations in 1884-1887 in both Siple and Dyer apparently are the result of two separate volcanic events. A similar volcanic doublet was found in an East Dronning Maud Land (G15) core [Moore et al., 1991] and in south pole cores [Delmas et al., 1992]. As Krakatoa ( $6^\circ\text{S}$ ,  $105^\circ\text{E}$ ) erupted in August 1883, it is likely that its  $\text{SO}_4^{2-}$  deposition in Antarctica would begin a year later (middle to late 1884). Indeed,  $\text{SO}_4^{2-}$  concentrations begin to rise above the seasonal background in the latter half of 1884 or early 1885, suggesting that the Krakatoa eruption is the likely source of S4 and D4. The  $\text{SO}_4^{2-}$  flux increase in late 1886 and 1887 (S3 and D3) is unlikely to be from Krakatoa, 3.5 years prior. Both Delmas et al. [1992] and Moore et al. [1991] suggested the June 1886 eruption of Tarawera ( $38^\circ\text{S}$ ,  $176^\circ\text{E}$ ) in New Zealand as the source of the second event in the doublet. Our precisely dated records support their conclusions. The short interval (less than 1 year) between the eruption and the bulk  $\text{SO}_4^{2-}$  deposition probably reflects the closer proximity of Tarawera to Antarctica. The assignment of the later event in the doublet to Tarawera is further supported by the lack of a contemporaneous event in Greenland.

**S5 (1836-37) and D5 (1836-38).** S5 and D5  $\text{SO}_4^{2-}$  deposit begins in 1836 and the small magnitudes are consistent with the January 1835 eruption of Cosiguina in Nicaragua ( $13^\circ\text{N}$ ,  $87^\circ\text{W}$ ; VEI=5), which may not have been a sulfur-rich eruption [Self et al., 1989]. The flux calculation shows that  $\text{SO}_4^{2-}$  deposition from Cosiguina lasted 3 years (1836-1838) at Dyer but only 2 years at Siple. Considering that Tambora lasted only 2 years, the 3-year deposition is unusual for Cosiguina. The third year (1838) at Dyer may reflect a higher-than-normal  $\text{SO}_4^{2-}$  background, leading to a possible overestimation of the net D5 volcanic  $\text{SO}_4^{2-}$  flux for Cosiguina.

**S6 (1832) and D6 (1834).** S6 and D6 are separated by 2 years. Given the high dating accuracy ( $\pm 0$  year) at this depth in the cores, it is impossible that these reflect the same event. D6 appears to result from high snow accumulation in 1834 (600 mm w.e.; the average for Dyer is 450 mm). Delmas et al. [1992] and Langway et al. [1995] reported a weak volcanic signal just a few years prior to Cosiguina (1836) in their respective cores, which they suspected to be the 1831 Babuyan ( $19.5^\circ\text{N}$ ,  $122.0^\circ\text{E}$ ) eruption in the Philippines.

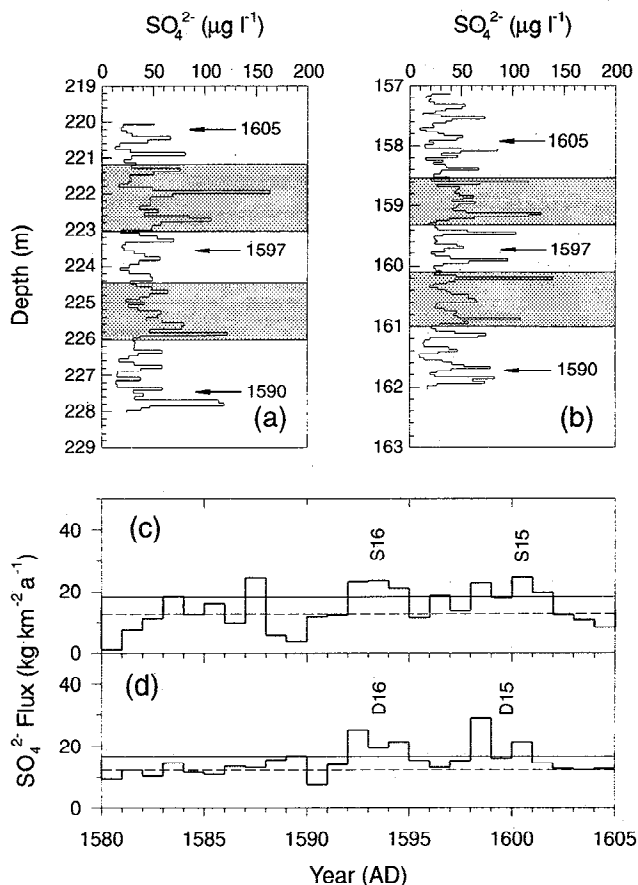
Although possible, the absence of a corresponding signal in Dyer casts doubt on the Babuyan eruption as the source for S6. It is probable that both S6 and D6 reflect high  $\text{SO}_4^{2-}$  flux from nonvolcanic sources.

**S7 (1816-17), D7 (1816-17) and S8 (1810-11), D8 (1810-11).** This well-known doublet has been found in all previous Antarctic ice cores. The younger event, dated at 1816 and 1817 in both cores, reflects the 1815 Tambora eruption. The 1-year lag between the eruption and the initial  $\text{SO}_4^{2-}$  deposition is consistent with the transport time of volcanic aerosols from the tropics.

With regard to event 8, no significant eruptions are found in the VEI records in the years immediately prior to Tambora. Comparison with the contemporaneous  $\text{SO}_4^{2-}$  doublet in Greenland cores led Dai *et al.* [1991] to attribute the older event to an unknown eruption in 1809 in the tropics. It is possible that a high-latitude SH eruption produced the 1810-1811 Antarctic signal [Moore *et al.*, 1991], and a coincidental eruption in the NH produced the 1810-1811 signal in Greenland. However, the close match between the timing of the 1809 event in Antarctic and Greenland cores and the similarity of the relative signal amplitudes in both polar regions support our original conclusion that a single, low-latitude eruption is the most likely source of the bipolar volcanic signals.

**S9 (1695-96), D9 (1696-97) and S10 (1693), D10 (1692).** The period from 1691 to 1697 contains elevated  $\text{SO}_4^{2-}$  concentrations and fluxes in both Dyer and Siple (Figure 4). Two events (eruptions) are identified using our selection method, and the possibility of more eruptions occurring within a 2 to 3-year period cannot be excluded. No volcanic eruptions have been identified in previous Antarctic ice cores during this period. In the GISP2 core, Zielinski *et al.* [1994] found volcanic signals in 1693 and 1695 and attributed them to NH eruptions (in Iceland and Japan). It is highly unlikely that those eruptions also produced the Antarctic signals of events 9 and 10 at Siple and Dyer. According to Simkin *et al.* [1981] a series of moderate (VEI=3) eruptions occurred in the Banda Sea area of Indonesia ( $\sim 7^\circ\text{S}$ ) during 1693 and 1694 and another moderate (VEI=3+) eruption of Reventador in Ecuador ( $8^\circ\text{S}$ ,  $78^\circ\text{W}$ ) in 1691. If they injected sulfur-rich aerosols directly into the stratosphere, these eruptions could have caused elevated  $\text{SO}_4^{2-}$  concentrations in the snow in both polar areas. However, the possibility cannot be discounted that moderate or large sub-Antarctic eruptions are responsible for events 9 and 10, since the historical records for these areas are rather poor [Newall and Self, 1982; Simkin *et al.*, 1981]. In any case, the accurately dated Siple and Dyer cores contain elevated  $\text{SO}_4^{2-}$  flux due to two or more volcanic eruptions in the period of 1690 to 1697.

**S11 (1673-74) and D11 (1673-74).** The years of 1673 and 1674 (dating uncertainty at this age:  $\pm 2$  years) is characterized by elevated  $\text{SO}_4^{2-}$  concentrations and fluxes at both Siple and Dyer (Figure 4). There are no previous ice core records of volcanic signals during this period. Such moderate events may have been easily missed in previous Antarctic ice cores, which were not analyzed for  $\text{SO}_4^{2-}$  continuously and with high sampling frequency. Newall and Self [1982] and Simkin *et al.* [1981] reported a moderately large (VEI=4?) eruption of San Salvador in El Salvador ( $14^\circ\text{N}$ ,  $89^\circ\text{W}$ ) in 1671 and a VEI 4 eruption in Indonesia (Gamkonora,  $1^\circ\text{N}$ ,  $127^\circ\text{E}$ ) in



**Figure 4.** Sulfate concentrations ( $\mu\text{g L}^{-1}$ ) are shown on a depth scale for the Siple (a) and Dyer (b) cores for the period from 1670 to 1700. The shaded areas indicate volcanic  $\text{SO}_4^{2-}$  deposit. (c and d) Annual  $\text{SO}_4^{2-}$  fluxes for Siple and Dyer, respectively, for the same time period.

May 1673. Either the San Salvador or both of these eruptions may have produced or contributed to event 11, although this cannot yet be confirmed.

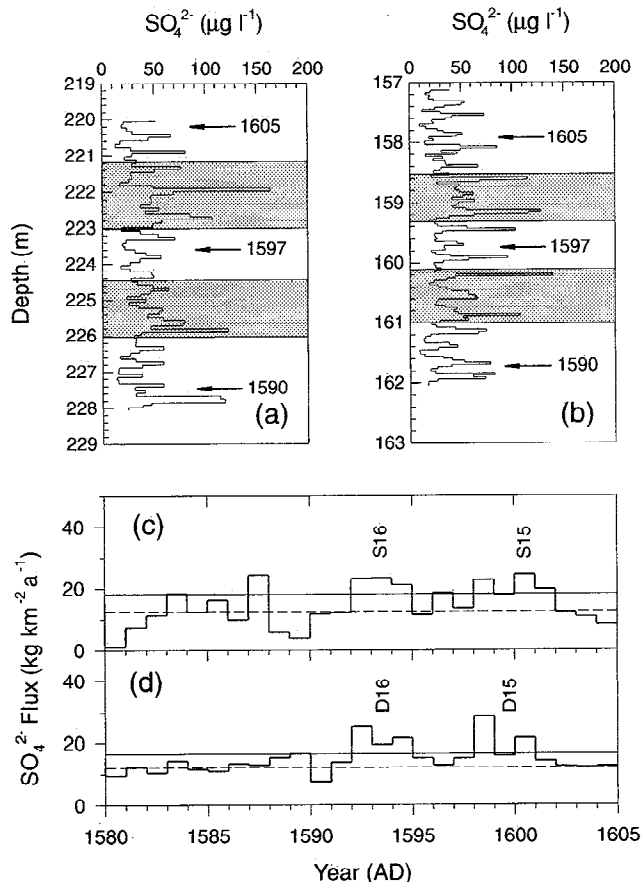
**S12 (1640-41) and D12 (1640-41).** This event appears to be a moderately strong eruption with a 2-year deposition pattern (Figure 3). Delmas *et al.* [1992] found a modest signal in the 1641 ice layer in a south pole core and suspected an Antarctic (Deception Island) eruption. There is a rather strong (VEI=5) eruption of Awu ( $4^\circ\text{S}$ ,  $125^\circ\text{E}$ ) in Indonesia in January 1641 [Simkin *et al.*, 1981], and a signal (1640) in the GISP2 core was attributed to this eruption [Zielinski *et al.*, 1994]. Such a strong tropical eruption should also leave a detectable signal in Antarctica. The probable years of  $\text{SO}_4^{2-}$  deposition from Awu (1641-1642) are within the time interval (1638-1643, dating uncertainty at this depth:  $\pm 2$  years) of event 12 in our records. The first year of deposition (1640) in the annual flux records appears to suggest that the Awu eruption may have occurred one or two years before the date reported by Simkin *et al.* [1981], given the same date (1640) of this event in the Siple and Dyer cores and in the GISP2 core. On the other hand, it is possible that we have overcounted the number of annual layers by 1 or 2 years. Our date of 1599 for the probable Huaynaputina eruption (1600) in these cores (discussed below) seems to verify the overcounting.



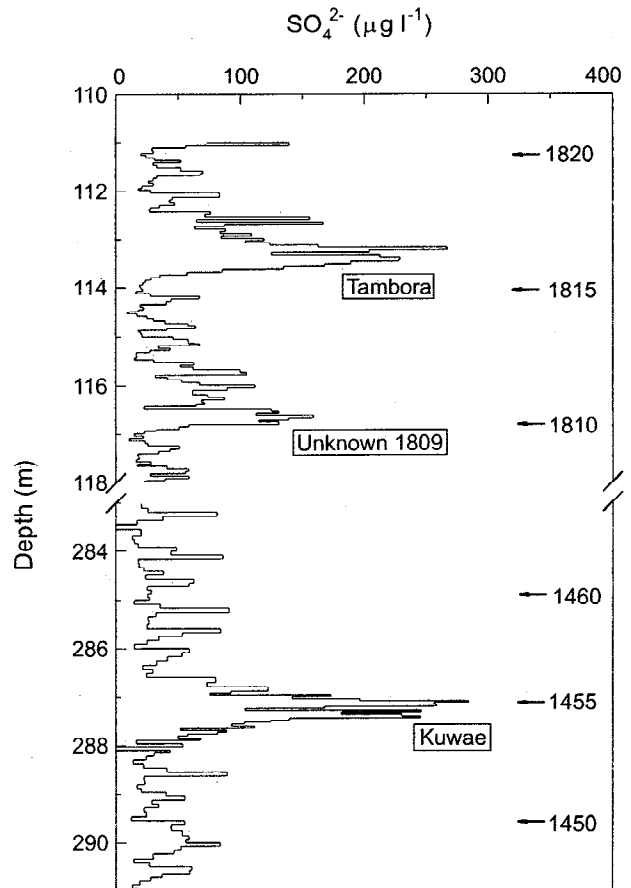
**S13 (1622).** No matching signal for S13 is found in Dyer and no records, including ice core records, of any significant volcanic eruptions exist. Although the high  $\text{SO}_4^{2-}$  flux results from the high summer  $\text{SO}_4^{2-}$  concentrations, rather than from a year of exceptionally high accumulation rate, it is unlikely that S13 reflects a volcanic event, given the lack of a corresponding signal in Dyer.

**S14 (1619) and D14 (1619-20).** This event is more prominent in Dyer than in Siple (Figure 3). The volcanic  $\text{SO}_4^{2-}$  deposition began in early 1619. *Delmas et al.* [1992] dated a moderate south pole signal at 1621 and attributed it to a sub-Antarctic eruption, based on the composition of particles found in ice. Our data and the lack of a corresponding signal in Greenland supports their suggestion of an eruption source somewhere in the middle to high southern latitudes. Although not identified as volcanic, a DEP/acidity signal also appears around 1620 in an East Antarctic core [see *Moore et al.*, 1991, Figure 3].

**S15 (1599-1602), D15 (1599-1601) and S16 (1593-95), D16 (1593-95).** Another pair of events is shown in Figure 5. Event 16 lasts 3 years (1593-1595) both at Siple and at Dyer, followed by event 15 starting in 1599. *Delmas et al.* [1992] and *Kirchner and Delmas* [1988] found a similar doublet in south pole cores during this decade, but the precise age difference between the two events could not be determined.



**Figure 5.** Sulfate concentrations ( $\mu\text{g L}^{-1}$ ) are shown on a depth scale for the Siple (a) and Dyer (b) cores for the period from 1590 to 1602. The shaded areas indicate volcanic  $\text{SO}_4^{2-}$  deposit. (c and d) Annual  $\text{SO}_4^{2-}$  fluxes for Siple and Dyer, respectively, for the same time period.



**Figure 6.** Highly elevated  $\text{SO}_4^{2-}$  concentrations caused by the Tambora (1815), the Unknown (1809), and the Kuwae (1453) eruptions are shown on a depth scale. The explosive character of the Kuwae eruption is similar to that of Tambora, considered the largest volcanic eruption in the last half millennium.

On the basis of elemental analyses of volcanic particles (tephra) found in the same cores, *Palais et al.* [1990] concluded that the younger event is from the 1600 eruption of Huaynaputina ( $17^\circ\text{S}$ ,  $71^\circ\text{W}$ ) in Peru. The age of event 15 suggests that it is from the Huaynaputina eruption. If so, our dating (1599) of this event is off by 1 or 2 years (the earliest possible deposition is late 1600), which is consistent with the overestimation of event 12 (Awu, 1640) but still within the dating range indicated by the counting errors ( $\pm 2$  years). *Palais et al.* [1990] also suggested that the older event in the south pole doublet may be from the 1595 eruption of Ruiz in Colombia ( $5^\circ\text{S}$ ,  $75^\circ\text{W}$ ), implying an age difference of 5 years between the two events (both eruptions occurred in the same season, March and February, of their respective years). However, the Siple and Dyer data clearly show that the age difference is 6 years between the initial  $\text{SO}_4^{2-}$  deposition of both events. As the confidence is high on the Huaynaputina identification [*Palais et al.*, 1990], the 6-year age difference casts doubt on Ruiz as the source for the older event, although the Ruiz eruption could be responsible for the volcanic deposit in the last 2 years of the 3-year event. If the younger event is indeed Huaynaputina, we suggest that the source of the volcanic  $\text{SO}_4^{2-}$  arriving in 1593 may be another eruption 1 or 2 years before Ruiz. A moderate (VEI=4)



eruption (Raung, Indonesia, 8°S, 114°E) is documented by *Newall and Self* [1982] and *Simkin et al.* [1981].

**S17 (1454-57).** One of the most interesting features in the Siple core is the section containing extremely high SO<sub>4</sub><sup>2-</sup> concentrations. This feature begins in 1454 and lasts 4 full years (Figure 6, bottom portion), representing a sustained and/or tremendously explosive eruption. Given the total ±3-year dating uncertainty at this depth, the data strongly suggest an eruption date between 1451 and 1456. *Pang* [1993] examined historic records from Europe and Asia and concluded that a very explosive eruption of Kuwae (17°S, 168°E) in the South Pacific occurred in early 1453, and its global scale atmospheric effects lasted at least through 1456. This conclusion is consistent with event S17 in the Siple record and the eruption date is verified by our accurately dated ice core records. A large volcanic event has been dated between 1450 and 1464 in various Antarctic ice cores [*Delmas et al.*, 1992; *Langway et al.*, 1995; *Moore et al.*, 1991]. *Zielinski et al.* [1994] reported a large eruption at 1459-1460 in GISP2 which was attributed to the Kuwae eruption. It is likely that all of these ice core volcanic signals originate from the explosive Kuwae eruption of 1453.

**S18 (1443).** Unfortunately, the Dyer SO<sub>4</sub><sup>2-</sup> record does not extend this far back in time. It is difficult to evaluate this moderate signal using only the Siple core. It is worth noting that no volcanic signal has been reported in any previous ice core records. S18 may be similar to S13, a year with exceptionally high SO<sub>4</sub><sup>2-</sup> background, due to unusually high accumulation for that year (650 mm w.e.; core average=550 mm w.e.). However, it is also possible that S18 is from a moderate eruption somewhere in the middle or high southern latitudes.

### Comparison of Volcanic Events

The net volcanic SO<sub>4</sub><sup>2-</sup> flux for each event (Table 2) is calculated as the difference between the sum of SO<sub>4</sub><sup>2-</sup> flux of the volcanic years and the average background SO<sub>4</sub><sup>2-</sup> flux. The volcanic SO<sub>4</sub><sup>2-</sup> flux, similar to volcanic acidity and the volcanic SO<sub>4</sub><sup>2-</sup> concentration, can be used to compare the magnitude of eruptions and to evaluate quantitatively the climatic impact of specific eruptions [*Robock and Free*, 1995; *Zielinski*, 1995]. However, it is not yet well understood how various air-to-snow mass transfer mechanisms (wet and dry deposition, riming, and vapor transfer) of atmospheric species such as SO<sub>4</sub><sup>2-</sup> affect the flux of volcanic SO<sub>4</sub><sup>2-</sup>. Therefore by using only the volcanic SO<sub>4</sub><sup>2-</sup> flux, it is difficult to compare volcanic events in ice cores obtained from various locations, as different mass transfer mechanisms may be dominant at different ice core locations. For example, the spatial variability for the Tambora H<sub>2</sub>SO<sub>4</sub> fluxes is quite large among the various ice cores from Greenland [*Clausen and Hammer*, 1988]. It is probably better to compare different events in the same ice core, as the depositional mechanisms are likely to be more consistent at the same location. One approach to comparing volcanic SO<sub>4</sub><sup>2-</sup> events among different ice cores is to first calculate the SO<sub>4</sub><sup>2-</sup> flux ratio of a specific event to that of a well-documented one in each ice core. The normalized values of that event can then be compared among the cores. For example, *Dai et al.* [1991] estimated the total SO<sub>2</sub> emission from the Unknown

**Table 2.** Volcanic SO<sub>4</sub><sup>2-</sup> Fluxes (*f*, in kilogram per square kilometer) for All Events in Siple and Dyer

Eruption	Siple		Dyer		<i>R<sub>S</sub></i> - <i>R<sub>D</sub></i> *
	Event	<i>f</i> / <i>ff<sub>Tambora</sub></i>	Event	<i>f</i> / <i>ff<sub>Tambora</sub></i>	
Agung 1963	S1	33 0.25	D1	13 0.15	+0.10
? (1937)	S2	11 0.08	D2	9 0.10	-0.02
Tarawera 1886	S3	12 0.09	D3	20 0.22	-0.13
Krakatoa 1883	S4	17 0.13	D4	11 0.12	+0.01
Cosiguina 1835	S5	26 0.19	D5	33 0.36	-0.17
Tambora 1815	S7	133 <b>1.00</b>	D7	90 <b>1.00</b>	<b>0.00</b>
Unknown 1809	S8	54 0.40	D8	54 0.60	-0.20
? (1695-1697)	S9	43 0.33	D9	24 0.27	+0.06
? (1692-1693)	S10	11 0.08	D10	16 0.17	-0.09
? (1673-1674)	S11	19 0.14	D11	22 0.25	-0.11
Awu 1641 and others	S12	34 0.25	D12	13 0.14	+0.11
? (1619-1620)	S14	16 0.12	D14	18 0.19	-0.07
Huaynaputina 1600	S15	34 0.26	D15	30 0.33	-0.07
? (1593-1595)	S16	30 0.22	D16	30 0.33	-0.11
Kuwae 1453	S17	122 0.92	--		
? (1443)	S18	13 0.10	--		

The year of eruption follows an identified event. Unknown eruptions are marked with a question mark, and their event dates are given in parentheses.

\* *R<sub>S</sub>*=*ff<sub>Tambora</sub>* for Siple, *R<sub>D</sub>*=*ff<sub>Tambora</sub>* for Dyer.

1809 eruption using the ratios of the volcanic SO<sub>4</sub><sup>2-</sup> flux of two events in several Greenland and Antarctic cores. Here the volcanic SO<sub>4</sub><sup>2-</sup> flux for each of the identified events is normalized against that of Tambora in their respective cores (Table 2). Data in Table 2 show that events 1 (Agung), 8 (Unknown 1809), 9 (1695), 12 (Awu?), 15 (Huaynaputina), and 16 (1593) are all moderately large eruptions producing volcanic SO<sub>4</sub><sup>2-</sup> flux of 25% to 50% that of Tambora. The Dyer results support our previous conclusion [*Dai et al.*, 1991] that the Unknown Eruption of 1809 eruption emitted approximately half the amount of SO<sub>2</sub> as Tambora.

The differences between the normalized flux of any specific event for Siple and Dyer (Table 2) are all within -0.20 to +0.20. This suggests that the core-to-core variability is within ±20% of the Tambora flux, when using such an approach to evaluate a specific event in Antarctic cores. However, this approach needs to be verified with additional ice core data sets.

Previous ice core volcanic chronologies show that Tambora is the most explosive eruption (in terms of volcanic SO<sub>4</sub><sup>2-</sup> aerosols produced) in the past 500-700 years. In Greenland, the Kuwae SO<sub>4</sub><sup>2-</sup> concentrations are lower than those from Tambora [*Zielinski et al.*, 1994], while in a Byrd Station (Antarctica) core, the Kuwae SO<sub>4</sub><sup>2-</sup> concentrations are higher than those from Tambora [*Langway et al.*, 1995]. In the Siple core, some individual samples associated with the Kuwae eruption contain SO<sub>4</sub><sup>2-</sup> concentrations higher than individual samples associated with the Tambora eruption

(Figure 6). In addition, the  $\text{SO}_4^{2-}$  deposition from Kuwae lasted longer (4 years) than that from Tambora (2 years). Nonetheless, net volcanic  $\text{SO}_4^{2-}$  flux from Kuwae ( $122 \text{ kg km}^{-2}$ ) is very similar to that for Tambora ( $133 \text{ kg km}^{-2}$ ). This is consistent with the estimate of magma volume (Dense Rock Equivalent (DRE)) erupted by Kuwae which is slightly less than that erupted by Tambora [Monzier *et al.*, 1994]. The Siple results show that the amount of  $\text{SO}_4^{2-}$  aerosols produced by the Kuwae eruption is comparable with that produced by Tambora. No records of any significant eruptions can be found during this time period (1450-1460) in either VEI or DVI. Since Kuwae is a low-latitude volcano and the signal of this eruption is found in Greenland ice cores, its atmospheric impact must have been global, and given its estimated DRE and  $\text{SO}_2$  emission, it clearly warrants a 6 or 7 designation on the VEI scale.

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