GEOPHYSICAL RESEARCH LETTERS, VOL. 24, NO. 21, PAGES 2679-2682, NOVEMBER 1, 1997

Quantifying the Pinatubo volcanic signal in south polar snow

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Abstract. Recent snow and firn core samples from South Pole contain increased sulfate (SO_4^{2-}) concentrations during 1992-1994 as a result of the June 1991 Pinatubo eruption and the August 1991 Cerro Hudson eruption in Chile. Traces of Pinatubo tephra (volcanic ash) were identified in the 1993 and 1994 snow layers, supporting the conclusion that increased SO_4^{2-} in 1993-1994 is from the Pinatubo eruption. Although the Pinatubo eruption preceded Hudson, its SO_4^{2-} signal in south polar snow follows and is resolved from that of Hudson. The deposition of the Pinatubo SO_4^{2-} aerosol was delayed due to the long transport to the high southern latitudes and its initial existence at high altitudes in the Antarctic atmosphere. Multi-year, multi-site sampling demonstrates that the volcanic signals are well preserved and spatially consistent. Measurements on 2 firn cores show that the South Pole SO_4^{2-} flux from Pinatubo is 10.9±1.2 kg km^{-2} over 2.2 years, while the Hudson flux is 3.2±0.6 kg km^{-2} in 1.1 years. These results, when combined with satellite-determined Pinatubo sulfur dioxide (SO_2) emission, make it possible to link quantitatively the atmospheric aerosol mass loading from a low-latitude volcanic eruption to its signal in polar ice cores.

Introduction

Volcanic eruptions, especially explosive low-latitude eruptions, introduce large amounts of sulfurous gases (mainly SO_2) into the global atmosphere. During their atmospheric lifetime of a few months to a few years, sulfuric acid/water aerosol particles can significantly alter the atmospheric albedo and therefore influence global climate. For example, following the June 1991 eruption of Mt. Pinatubo (15.14°N, 120.35°E), a decrease of 0.2-0.7°C was observed in global tropospheric and near-surface temperatures [*Jones and Kelly*, 1996; *McCormick et al.*, 1995], although winter warming occurred over areas of large landmass [*Robock and Mao*, 1995].

The 1991 Pinatubo eruption was monitored by both airborne and satellite instruments [e.g., *McCromick and Veiga*, 1992]. Sulfur-rich eruption clouds reached 25-35 km and contained an estimated total of 18±2 million tons (1 Mt=1 Tg=10^{12} g) of SO_2 [*Krueger et al.*, 1995]. Within two months the SO_2 in the stratosphere was oxidized [*Bekki et al.*, 1993] to a total aerosol loading of 28 Mt sulfuric acid. The Pinatubo aerosol mass began to spread poleward in late August or early September [*Trepte et al.*, 1993] and, by mid-1992, covered the entire surface of the globe [*Hitchman et al.*, 1994]. Lidar measurements at McMurdo and South Pole Stations in Antarctica first detected Pinatubo aerosols in late 1991 [*Cacciani et al.*, 1993; *Deshler et al.*, 1994].

Volcanic SO_4^{2-} aerosols, along with fine tephra (volcanic ash), are gradually removed from the atmosphere and the aerosol deposition results in increased SO_4^{2-} concentrations in polar snow. Chemical identification of tephra can be used to link increased SO_4^{2-} levels in ice cores to specific volcanoes [e.g., *Palais et al.*, 1990]. The removal of Pinatubo aerosols was expected to produce a significant increase of SO_4^{2-} concentrations in post-1991 Antarctic snow. Indeed, *Dibb and Whitlow* [1996] report finding increased SO_4^{2-} in several snowpits excavated at South Pole in 1994 and attribute the increase to the Pinatubo eruption with possible contributions from the August 1991 eruption of Cerro Hudson in Chile (45.92°S, 73.00°W).

Volcanic acidity and SO_4^{2-} in polar ice cores are used to construct the history of past volcanism [*Delmas et al.*, 1992; *Zielinski et al.*, 1996]. High resolution, well-dated ice core records provide a unique tool to study the climate-volcanism connection [*Crowley et al.*, 1993; *Zielinski*, 1995]. However, atmospheric aerosol loadings from volcanic eruptions are not directly available from ice core records which only provide a relative measurement. Knowledge of the aerosol loadings is important for assessing the climatic impact of volcanic eruptions. The Pinatubo eruption offers a unique opportunity to study the quantitative relationship between the atmospheric aerosol loading from an explosive eruption and its SO_4^{2-} signal in polar ice cores. However, the Pinatubo signal in Antarctic snow may be coincidentally mixed with the fallout from the Hudson eruption, as suggested by *Dibb and Whitlow* [1996]. In this paper, we report the detection of both the Pinatubo and Hudson signals and describe the resolution of these two eruptions in South Pole snow in order to quantify the Pinatubo signal.

Snow Sampling and Analysis

In November 1993, two 0.80 m snowpits, located 20 km upwind (Grid 045 and 350) from the Amundsen-Scott South Pole Station, were excavated and vertically continuous snow samples were taken from pit walls with a clean plastic spatula and placed in pre-cleaned plastic containers. In December 1994, 6 shallow (20 meters) firn cores were drilled at distances of 9 to 11 km from the station along 6 lines of a snow accumulation network (see description below). The pit samples and cores were transported frozen to the Byrd Polar Research Center (BPRC) at The Ohio State University for later analysis. Concentrations of cations (Na^+, K^+, Mg^{2+} and Ca^{2+}) and anions (Cl^-, NO_3^- and SO_4^{2-}) were measured in the pit samples and in two of the six 1994 cores (Cores B and D; the other 4 cores were used for other analyses). A detailed description of the core handling and analytical procedures is given in *Dai et al.* [1995].

Also in December 1994, snow blocks were taken from a pit at 20 km from the South Pole Station down to the depth of 0.90 m. These snow blocks were melted and poured into pre-cleaned high density polyethylene bottles at the station. In the BPRC lab, insoluble particles in the meltwater were filtered onto 0.2 μm Millipore cellulose membrane filters. The filters were mounted on aluminum stubs, coated with evaporated carbon and subsequently examined using a JEOL JSM-820 scanning electron microscope (SEM). Quantitative analysis for the major element composition of particles on the filters was achieved with energy dispersive x-ray spectroscopy, using standard ZAF correction techniques.

Results

All SO_4^{2-} data are based on non-sea-salt (nss) SO_4^{2-}, as calculated from total SO_4^{2-} and Na^+ concentrations in snow. However, using total SO_4^{2-} rather than nss-SO_4^{2-} would not change the conclusions of this study, as nss-SO_4^{2-} constitutes 96% of the total SO_4^{2-}.

Vertical profiles of SO_4^{2-} concentrations in the top 2 m of the two 1994 firn cores are illustrated in Figure 1. *Dibb and Whitlow* [1996]

Paper number 97GL02734.
0094-8534/97/97GL-02734$05.00

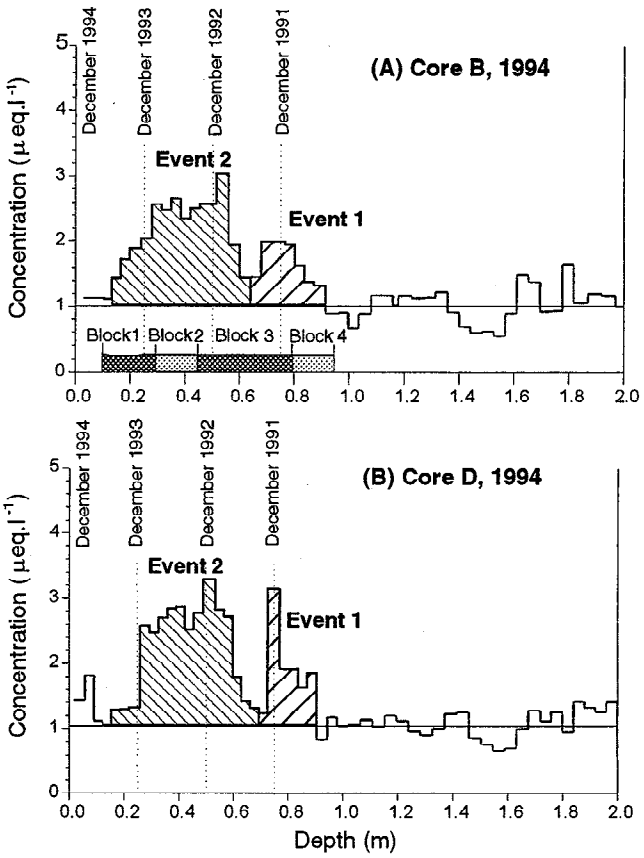

Figure 1. Nss-SO_4^{2-} concentrations in **(A)** Core B and **(B)** Core D. Two volcanic events are indicated by the shaded areas. Snow block samples (Blocks 1-4), marked on the x-axis of **(A)** were taken in a nearby pit according to their depth intervals. Horizontal lines indicate the non-volcanic SO_4^{2-} background. The December date lines are estimated from mean annual net snow accumulation at South Pole during 1992-1996.

used seasonal variations in the concentrations of Na^+, NO_3^- and the Cl^-/Na^+ ratio to date South Pole snowpits. However, among the measured ion concentrations and $\delta^{18}O$, we found no parameters that exhibited unambiguous seasonal variations. This may be due to our lower sampling frequency (5 samples/year) than that used by *Dibb and Whitlow* (~10 samples/year). Therefore we dated the cores assuming a constant annual net snow accumulation rate. A 236-pole snow accumulation network, extending 20 km from the South Pole Station in 6 directions (Grid 045, 110, 170, 230, and 350), was established in 1992 and annual accumulation measurements have been made since then. The spatially averaged mean annual net snow accumulation of 0.23±0.09 m from this network for 1992-1996 is used in dating the cores. The top 2 m of the firn cores are dated as follows: the snow surface is December 1994, the December lines for the previous 3 years (1993, 1992 and 1991) are identified at 0.23, 0.46 and 0.69 m below the surface, respectively; mid-winter (June) snow is assumed to be midway between 2 adjacent December lines. The 1993 snowpits (Figure 2) are dated using the same method. The large spatial variability (±0.09 m/year) associated with the mean annual net snow accumulation implies that the time lines given in this paper carry an uncertainty of as much as 40%.

The most remarkable feature in Figure 1 is the marked increase of SO_4^{2-} concentrations above 0.90 m, approximately late 1991. Prior to the Pinatubo eruption was a period of volcanic quiescence [*Hitchman et al.*, 1994] and the low SO_4^{2-} concentrations below 0.90 m are considered representative of the non-volcanic background. The SO_4^{2-} concentrations declined sharply in early to mid-1994 and returned to non-volcanic background levels (Figure 1). In the 1993 snowpits, SO_4^{2-} increased similarly beginning at 0.7 m (Figure 2) and remained high at the time when the pit samples were taken (November 1993).

Several tephra particles with chemical composition similar to that of Pinatubo ash were found in the 1994 snow blocks at depths between 0.10-0.44 m (Blocks 1 and 2 in Figure 1), representing the time period of mid-1994 to early 1993. No tephra similar to that of Pinatubo was found below 0.44 m or prior to early 1993.

Discussion

Volcanic Events During 1991-1994

In Figure 1, two "events" (shaded areas) of increased SO_4^{2-} are identified in both cores. A relatively narrow "peak" (Event 1) above the extrapolated background occurs between 0.65-0.90 m. According to the dating, this depth interval represents snowfall between mid-1991 and mid-1992. Event 2, a broad, more sustained increase, follows Event 1 and is dated between mid-1992 and mid-1994. The 2 snowpits sampled a year earlier show 2 similar events (Figure 2). Event 1 in Pit 1 appears to predate mid-1991 when Pinatubo and Hudson erupted. However only non-volcanic background SO_4^{2-} concentrations are possible in pre-1991 snow. A specific volcanic event exists in the same snow layer in all pits and cores. Therefore, Event 1 in Pit 1 is likely the same as Event 1 in Cores B and D but is buried deeper than expected due to high snow accumulation at this pit location.

The high SO_4^{2-} concentrations in Figures 1 and 2 are consistent with the prominent SO_4^{2-} spike *Dibb and Whitlow* [1996] found in

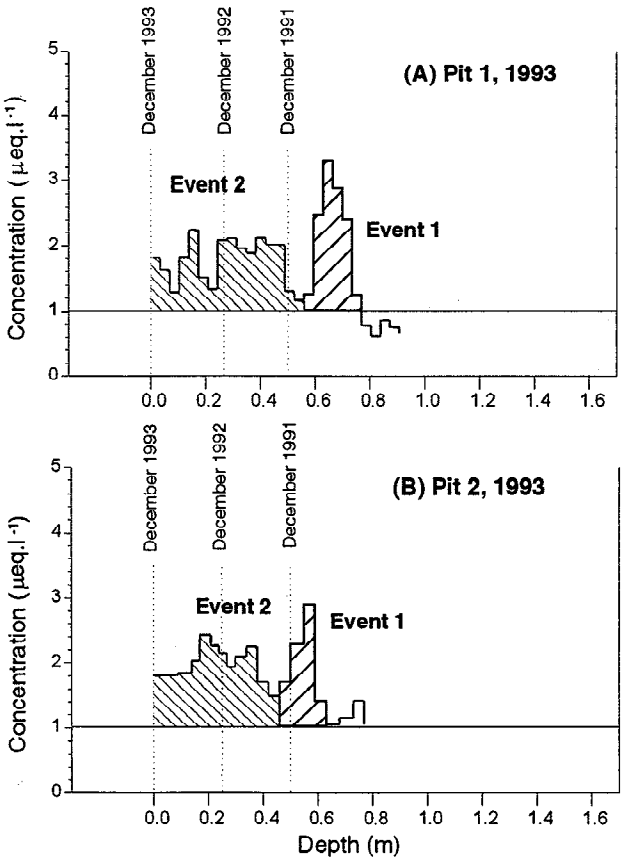

Figure 2. Nss-SO_4^{2-} concentrations in two snowpits excavated in November 1993 at South Pole. Horizontal lines represent the non-volcanic background. Date lines are estimated from mean annual net snow accumulation, as in Figure 1. Event depths are shallower than in Figure 1 due to snow accumulated during 1994.

3 South Pole snowpits. They concluded that the spike reflects the fallout from Pinatubo, with possible contributions from the Hudson eruption. We discuss below the possible influence of the Hudson eruption on the Pinatubo signal in South Pole snow and attempt to quantify the Pinatubo signal.

The Hudson Signal in Snow

The August 1991 Hudson eruption emitted an estimated 1.5 Mt SO_2 into the atmosphere [*Doiron et al.*, 1991]. Although the SO_2 output from Hudson is less than 10% that of Pinatubo, significant deposition of Hudson SO_4^{2-} in Antarctic snow is likely, due to Hudson's relative proximity to Antarctica and the rapid transport of its aerosols into the high southern latitudes [*Doiron et al.*, 1991]. The close timing of the two eruptions and the longer transport time for Pinatubo aerosols to reach Antarctica may result in simultaneous deposition of their SO_4^{2-} aerosols in Antarctic snow. This would make it difficult to quantify the Pinatubo signal.

However, satellite observations [*Schoeberl et al.*, 1993] show that the Hudson cloud began to move southward immediately after the eruption in mid-August, 1991, when the bulk of the Pinatubo cloud was still confined to the equatorial atmosphere. The Hudson aerosol mass was clearly present in the lower polar stratosphere during the 1991 austral spring [*Deshler et al.*, 1992], whereas the front of the Pinatubo aerosol arrived after the breakup of the 1991 Antarctic polar vortex [*Trepte et al.*, 1993]. Satellite (SAGE II and NOAA/AVHRR) observations [*Herber et al.*, 1996; *Long and Stowe*, 1994] also show that the bulk of the Pinatubo aerosol mass did not move into the high southern latitudes during the first six months after the eruption. Furthermore, lidar monitoring of stratospheric aerosols at South Pole from September 1991 to June 1992 clearly showed that two distinctly separate aerosol layers existed in the polar stratosphere [*Cacciani et al.*, 1993]. The lower layer was attributed to Hudson and its disappearance from the stratosphere was observed by the end of December 1991 [*Cacciani et al.*, 1993]. The rapid disappearance of the Hudson aerosol layer may have been facilitated by the enhanced stratosphere-to-troposphere exchange associated with the 1991 polar vortex. Once in the troposphere, the volcanic aerosol particles are rapidly removed through precipitation and other depositional processes. The upper aerosol layer, attributed to Pinatubo, was first observed in December 1991. The arrival of the Pinatubo aerosol mass in late spring or summer, after the breakup of the 1991 polar vortex, may have effectively prevented its mixing with the lower aerosol layer. In fact, the center of this upper layer remained above the tropopause well into 1992 [*Cacciani et al.*, 1993], and a substantially large portion of the aerosol mass was observed in the 1992 polar vortex. Given these atmospheric observations, the Hudson SO_4^{2-} signal is likely to be in the snow layer of late 1991 to early 1992 and to be of short duration. The Pinatubo signal, on the other hand, is expected to be a strong and broad peak starting in mid-1992 at the earliest. Such a scenario appears to be consistent with the 2 events in Figures 1 and 2. This led us to conclude that Event 1 in the snowpits and firn cores is derived from the Hudson SO_4^{2-} aerosol, and that Event 2 represents the fallout from Pinatubo. The low (close to the non-volcanic background) SO_4^{2-} concentrations of the samples at about 65 cm in both cores (Figure 1) appear to indicate only a minimal overlap between the 2 events.

The results of the tephra search in the 1994 snow block samples support the identification of Event 2 as Pinatubo. Several volcanic particles (2-10 μm) with a chemical composition matching that of Pinatubo glass (Table 1) were found between early 1993 and mid-1994 (Blocks 1 and 2 in Figure 1(A)), but not earlier (Blocks 3 and 4). The absence of Pinatubo tephra at the depth interval of Event 1 indicates, although not conclusively, that Event 1 is not associated with Pinatubo. The failure to find Pinatubo tephra in Block 3 which covered part of Event 2 may be due to the fact that there were too few tephra particles in the sample; or, alternatively, Block 3 may not include any part of Event 2 because snow accumulation was possibly higher than average at the site where the block samples were obtained.

Table 1. Composition of volcanic glass found in 1993 and 1994 snow samples (Blocks 1 and 2 in Figure 1(A)) is compared with that of Pinatubo ash. Pinatubo ash samples were collected at several locations (footnotes). Results are percentages of individual oxides determined by SEM/Energy dispersive x-ray spectroscopic analysis (instrumental precision ≈0.5%). "n" is the number of analyses/particles and standard deviations are included in parentheses.

	Location of collected Pinatubo ash samples					
	Particles on filters n=10	Near Pinatubo* (this work) n=16	South China Sea 1 n=6	South China Sea 2 n=14	Luzon Island 3 n=6	Luzon Island 4 n=16
SiO_2	77.35 (0.62)	76.68 (0.90)	77.14	77.5	76.55 (0.78)	73.69 (3.27)
TiO_2	0.23 (0.12)	0.18 (0.08)	0.12	0.12	0.06 (0.03)	0.16 (0.13)
Al_2O	12.56 (0.25)	12.87 (0.41)	12.96	12.96	12.47 (0.23)	13.34 (1.26)
FeO	1.13 (0.18)	1.13 (0.38)	0.81	0.79	0.75 (0.03)	1.39 (0.87)
MnO	0.08 (0.05)	0.05 (0.04)				
MgO	0.70 (0.15)	0.58 (0.10)	0.22	0.20	0.10 (0.02)	0.62 (0.72)
CaO	1.52 (0.19)	1.72 (0.36)	1.15	1.15	1.20 (0.04)	2.22 (1.50)
Na_2	2.56 (0.42)	3.02 (0.38)	2.26	2.08	3.52 (0.72)	2.85 (0.55)
K_2O	3.88 (0.34)	3.77 (0.37)	2.77	2.67	3.07 (0.24)	2.80 (0.48)
Total	100.00	100.00	97.43	97.47	97.11	97.08

*: fine ash (co-ignimbrite from the June 15, 1991 paroxysmal eruption) collected 15 km east of the Pinatubo vent [provided by *W.E. Scott*, USGS].

1 and 2: SCS-C91-S6 and SCS-C91-D9, glass shards in tephra fallout captured by sediment traps in the South China Sea [*Wiesner et al.*, 1995]; no standard deviations were given.

3 and 4: EW910615-1 and EW910615-2, glass in crystal-rich pumice and crystal-poor pumice, respectively, collected near the volcano [*Pallister et al.*, 1992].

Non-volcanic Background SO_4^{2-}

Non-volcanic SO_4^{2-} in Antarctic snow is derived from marine biogenic sulfur emissions (e.g., dimethyl sulfide or DMS). Detecting and estimating volcanic signals depends on how this variable background is determined [*Cole-Dai et al.*, 1997]. It has been suggested [*Legrand and Feniet-Saigne*, 1991] that methanesulfonic acid (MSA), an oxidation derivative of DMS, in Antarctic snow may be related to variations of El Niño. Based on the assumption that MSA and non-volcanic SO_4^{2-} are similarly affected by El Niño events, *Dibb and Whitlow* [1996] estimated the non-volcanic SO_4^{2-} background in recent South Pole snow and concluded that the Pinatubo SO_4^{2-} signal is rather small. However, it has not been demonstrated that non-volcanic SO_4^{2-} and MSA in Antarctic and South Pole snow are significantly correlated. In fact, in the same South Pole core in which MSA was suspected to be related to El Niño, *Legrand and Feniet-Saigne* [1991] reported no correlation between SO_4^{2-} and El Niño events. Similarly, *Pasteur et al.* [1995] found no significant link between either MSA or nss-SO_4^{2-} and El Niño events in an ice core from the Antarctic Peninsula region, near the source areas of DMS emissions. We therefore assume that the non-volcanic SO_4^{2-} background is a constant equal to the average concentration prior to Event 1. For Cores B and D the background is 1.01±0.11 and 1.05±0.09 μeq.l^{-1}, respectively.

Calculation of Net Pinatubo and Hudson SO_4^{2-} Flux

The net volcanic SO_4^{2-} flux (total flux minus background flux) for Pinatubo is calculated for Cores B and D as follows. The SO_4^{2-} flux of an individual sample is the product of its SO_4^{2-} concentration and the sample length in water equivalent. The Event 2 length is from the end of Event 1 (0.65 m) to when the SO_4^{2-} concentration falls to background levels (at 0.10 m in Core B and 0.15 cm in Core D) and the total flux is equal to the sum of the fluxes of samples within the event. Since deposition appears to be incomplete at the time when the 1993 pits were excavated, no flux calculation was

made from the pit data. The net Pinatubo SO_4^{2-} flux thus calculated is 10.9±1.2 and 10.9±0.9 kg km^{-2} for Cores B and D, respectively, with the uncertainties estimated from the non-volcanic background variability. Based on the 0.23 m/year snow accumulation rate, the duration of Pinatubo SO_4^{2-} deposition is estimated to be 2.2 years. The Hudson (Event 1) flux is similarly calculated (2.9±0.6 and 3.6±0.6 kg km^{-2}, respectively for Cores B and D), with an estimated duration of 1.1 years. While the total Hudson SO_2 emission is less than 10% of the Pinatubo output, its flux is about one third that of Pinatubo. This illustrates that the eruption of a volcano in mid- or high southern latitudes will result in an enhanced SO_4^{2-} signal, relative to a low latitude eruption, in Antarctic snow. The Pinatubo flux is comparable to those of the 1963 Agung eruption (9.1 kg km^{-2}, 1.7 years) and the 1883 Krakatoa eruption (8.7 kg km^{-2}, 2.7 years) reported by *Delmas et al.* [1992].

Conclusions

We found increased SO_4^{2-} concentrations in 1992-1994 South Pole snow, resulting from the 1991 eruptions of Pinatubo and Hudson volcanoes. Satellite and lidar observations that acrosol clouds from the eruptions existed at separate altitudes in the Antarctic atmosphere suggest that their SO_4^{2-} signals in Antarctic snow are likely to be segregated from each other, with the Hudson signal preceding that of Pinatubo. Two separate volcanic events are discernible and we conclude, with supporting tephra evidence, that the more recent event is from Pinatubo. Calculation of the net volcanic fluxes for Pinatubo and Hudson demonstrates that the Pinatubo eruption produced a moderate volcanic signal and that the signal from the much smaller Hudson eruption is enhanced relative to the Pinatubo signal, due to the Southern Hemisphere location of Hudson. It is now possible to combine the calculated Pinatubo SO_4^{2-} flux with the satellite-estimated SO_2 emission to investigate the relationship between the atmospheric aerosol loading by a low-latitude volcanic eruption and its SO_4^{2-} flux in polar snow. This will allow the quantitative estimation of atmospheric aerosol mass loadings of past volcanic eruptions recorded in polar ice cores.

Acknowledgments. We thank the Polar Ice Coring Office for successful retrieval of firn cores. W.E. Scott of the US Geological Survey generously provided Pinatubo ash samples and G.A. Zielinski provided valuable comments and suggestions on the tephra results. Thoughtful reviews by A. Robock and G.A. Zielinski improved the manuscript. This work was supported by NSF Grants OPP-9321478 and OPP-9526725 to The Ohio State University. This is Contribution Number 1053 of the Byrd Polar Research Center.

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(Received May 21, 1997; revised August 6, 1997;
accepted September 18, 1997.)