High Resolution Ice Core Records of Late Holocene Volcanism: Current and Future Contributions from the Greenland PARCA Cores

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A suite of spatially distributed, multi-century cores collected since 1995 under NASA's Program for Arctic Regional Climate Assessment (PARCA) provides an excellent archive of volcanic emissions reaching Greenland. As records of equivalent quality from higher accumulation sites in Antarctica become available, their integration will produce a richer, better temporally constrained and more climatologically valuable history of global volcanism. The Greenland PARCA cores have been accurately dated using multiple seasonally varying indicators ($\delta^{18}O$, insoluble dust, H_2O_2, nitrate, calcium) and the ongoing chemical analyses are providing new volcanic histories that complement the limited records that exist. The first results confirm that the sulfate aerosols from an unidentified pre-Tambora eruption called Unknown: (1) were widely dispersed across the Greenland ice sheet; (2) first arrived in the 1810 A.D. snow fall; and, (3) in 1810 A.D., the first year after the eruption (1809 A.D.), produced concentrations of excess SO_4^{2-} (EXS) comparable to those deposited in 1816 A.D., the first year after the eruption of Tambora in 1815 A.D. The EXS originating from the eruption of Laki craters or Lakagígar (1783 A.D.) is confined to a single year (1783 A.D.) and varies considerably across the ice sheet, primarily as a function of the local accumulation rate. Future chemical analyses of the PARCA cores promise richly detailed histories of EXS emissions from both known and yet to be identified volcanic eruptions. The high temporal resolution of these ice core records will help resolve timing issues and their broad spatial distribution will provide a more representative estimate of the EXS flux associated with a specific eruption.

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Volcanism and the Earth's Atmosphere
Geophysical Monograph 139

10.1029/139GM09

1. INTRODUCTION

The likelihood of anthropogenic climate forcing on a global scale has necessitated efforts to differentiate the contributions of natural forcing mechanisms from those attributable to human activities of the past two centuries. One natural cause of short-term climate change is the perturbation of the Earth's radiation balance by the emission of volcanic gases into the stratosphere. Here the aerosols may remain for several years, perturbing the earth-atmosphere radiation balance and hence the surface temperature distribution that modifies atmospheric and oceanic circulation

patterns. Papers in this volume review the character of both modern and ancient volcanic eruptions, their radiative effects, and efforts to measure and model the earth-system responses. The impacts of volcanism on the Earth's climate system are reviewed by *Robock* [2000] who stresses that evaluation of the causes of climatic change over the last few millennia requires a 'reliable' record of atmospheric aerosol loading from volcanic eruptions. This paper presents the first volcanic records from the Greenland PARCA cores that will eventually yield a rich and detailed history of atmospheric aerosols of volcanic origin.

No perfect archive of explosive volcanic eruptions exists as each recording system (e.g., ice cores, tree rings, documentary evidence) has its unique biases, quality of signal preservation, and dating difficulties. The low background concentration of most chemical species in polar ice cores makes them an excellent medium for identification of volcanic emissions. Holocene volcanic eruptions are rarely recorded as visible tephra layers in polar cores although a few have been reported [*Zielinski et al.*, 1997]. More generally volcanic events are identified by elevated concentrations of EXS deposited as particulate aerosols that form when gaseous compounds (mainly sulfur dioxide) are oxidized to sulfuric acid and water. If the gases are injected into the stratosphere, particularly from low to mid-latitude volcanoes, the aerosols can spread globally. Observations after the June 15, 1991 eruption of Mt. Pinatubo in the Philippines (15°8'N; 120°21'E) revealed that the aerosols rapidly spread zonally [*Bluth et al.*, 1992] and then began to disperse poleward [*Trepte et al.*, 1993].

In the last decade several authors have used ice cores to establish more firmly the history of explosive volcanism and to quantify the sulfate emissions attributable to specific eruptions for inclusion in climate models [*Robock and Free,* 1995; *Zielinski, 1995; Claussen et al., 1997; Crowley and Kim*, 1999; *Crowley*, 2000]. This paper highlights the valuable information that the high resolution PARCA cores can provide about the history of volcanic activity and the quantity of volcanic sulfate deposited over the Greenland ice sheet. The latter allows the associated stratospheric sulfate burden to be estimated and this is critical for including the effect of volcanic eruptions in climate models. The paper does not provide a comprehensive review of the Earth's volcanic history as reconstructed from ice cores, but it attempts to provide a balanced discussion of the strengths and limitations of ice core-derived volcanic information. Here we report new analyses that complement and expand previous volcanic chronologies and emissions estimates, confirm that eruption of the pre-Tambora Unknown event occurred in 1809 A.D., and provide new evidence suggesting that the sulfate emissions from Lakagígar (henceforth Laki) arrived on the Greenland ice sheet in 1783 and not in 1784.

2. ICE CORE-DERIVED VOLCANIC HISTORIES

Since the 1960s when the first long ice cores were recovered from Greenland (1966 at Camp Century) and Antarctica (1968 at Byrd Station), polar firn and ice cores have provided unique details about the nature, timing and impacts of volcanic eruptions on regional to global scales. *Gow and Williamson* [1971] reported numerous ash and dusty layers in Byrd ice core sections from the latter part of the Last Glacial Stage (LGS, ~16–30 ka BP). This lead to renewed interest in the possible role of prolonged, explosive volcanism on the climate at the Last Glacial Maximum (~18 ka BP) [*Harvey,* 1988]. Was the volcanism responsible for the occurrence of the coldest global temperatures just prior to climate recovery or did the volcanism extend the duration of the LGS? Subsequently, most of these events were attributed to regional (Antarctic or sub-Antarctic islands) eruptions [*Kyle et al.*, 1981] that likely did not affect the transparency of the global atmosphere. Nevertheless, renewed interest in the volcano-climate connection was generated and soon thereafter Hammer and his colleagues used acidity measurements to demonstrate that concentrations of sulfuric acid (H_2SO_4) in Greenland ice cores were elevated by deposition of volcanic aerosols [*Hammer*, 1977; *Hammer et al.,* 1980].

2.1. Dating Ice Cores

For three decades volcanic signals preserved in ice cores have been exploited as stratigraphic markers for dating ice cores [*Hammer et al.*, 1978; *Hammer*, 1989] and as archives for augmenting and extending the historical record of globally significant volcanic events [*Hammer*, 1980; *Delmas et al.*, 1985; *Dai et al.*, 1991; *Cole-Dai et al.*, 1997; 2000; *Zielinski*, 2000]. Precise dating of a proxy record is critical for meaningful interpretation and robust comparisons with other histories. The use of volcanic horizons as "time stratigraphic" markers for dating ice cores has created an unintended 'chicken and egg' situation. If the ice core sulfate deposit is used as a marker horizon for dating then it can no longer be used to independently ascertain the arrival time of the aerosols to the ice sheet. Volcanic layers can only be used to determine the timing of aerosol deposition when the horizon is dated by counting multiple seasonally varying constituents from some other independently confirmed event. Moreover, due to the potential for mixing of snow between consecutive years by blowing, drifting, deflation and redepo-

sition, the timing of an event should be confirmed in more than one core from a region [*Gow*, 1965; *Mosley-Thompson et al.*, 1985, 2001; *Pomeroy and Jones*, 1996; *Zielinski*, 2000]. The chance of snow layer mixing increases as annual net accumulation of snow at the site decreases.

Many of the cores collected in the 1970s and early 1980s were analyzed continuously by cutting discrete samples over the entire length of the core, but often the analyses were limited to the oxygen and/or hydrogen isotopic ratios ($\delta^{18}O$, δD, respectively). Although these species often show a seasonal signal they are also subject to post-depositional modification by vapor diffusion within the firn as well as by melting or percolation. When vague seasonal signals hinder layer counting efforts, other methods may be employed to augment the dating effort. One approach is to construct a simple depth-age model for the core using the annual accumulation, thickness of the ice sheet, and ice flow characteristics. Of course, this includes the unrealistic assumption of a steady state accumulation regime, but does provide an approximate time scale. With the approximate time scale in hand, known volcanic events can be "sought" by analyzing selected sections of core for acidity signals using electrical conductivity measurements (ECM) or by chemically analyzing samples to isolate layers containing elevated sulfate concentrations. This can be risky for ice core sections with closely spaced events that might be confused if only one event is detected. Even when cores are continuously analyzed by ECM, less prominent (or unanticipated) volcanic events can be missed. Although great improvements have been made in the use of rapid, *in situ*, nondestructive ECM analysis [*Taylor et al.*, 1997; *Wolff et al.*, 1997], the technique remains dependent upon core quality, temperature of the ice at the time the measurement is made, and operator experience. Ion chromatographic analysis of discrete samples, cut to discern annual cycles ($\approx$ 6 to 10 samples per accumulation year), is more time consuming and also dependent upon core quality, but can still be used when core quality is degraded and thus offers a more thorough examination of the core as well as more quantitative information about sulfate deposition. Additionally, as other non-volcanic chemical species may render the ice acidic, ion chromatographic analysis of sulfate (SO_4^{2-}) provides a more definitive indication of a volcanic emission and allows a more quantitative assessment of the amount of volcanic sulfate aerosol preserved in the ice.

2.2 Refining Volcanic Chronologies: Annual Resolution

Recent advances such as the continuous flow analysis (CFA) system [*Fuhrer et al.*, 1993; *Anklin et al.*, 1998] and improvements to ion chromatographic techniques now facilitate the analysis of smaller sample volumes. This has made it possible to use 'annual layer counting' in regions with lower annual average accumulation (100–200 mm a^{-1} water equivalent, w.e.) and has resulted in more continuous analyses so that the entire core length is chemically and/or physically characterized with high temporal resolution. The result is that more volcanic events, some known and some unknown, are now being identified, quantified and compared among multiple cores.

Ice cores collected to 303-meters depth in 1985 at Siple Station (75°55'S; 84°15'W) and to 235-meters depth in 1989/90 on the Dyer Plateau (70°40'S; 64°52'W) were among the first Antarctic cores to be analyzed continuously (top to bottom) for major anions (including SO_4^{2-}), $\delta^{18}O$, and insoluble dust. The high annual average accumulation at these sites (Siple: 560 mm a^{-1} w.e.; Dyer Plateau: 440 mm a^{-1} w.e.) made it possible to date the cores very accurately using the excellently preserved seasonal variations in both $\delta^{18}O$ and SO_4^{2-} [*Mosley-Thompson et al.*, 1991; *Mosley-Thompson*, 1992; *Thompson et al.*, 1994]. The resulting volcanic chronology from 1417 to 1989 A.D. resolved a number of temporal ambiguities regarding known eruptions and revealed some previously unknown events in the 1600s [*Cole-Dai et al.*, 1997]. The three largest eruptions, in terms of the sulfate flux deposited at these coring sites (assumed to reflect input to the Southern Hemisphere stratosphere) are Tambora in Indonesia (1815 A.D.), Kuwae ($\approx$ 1453 A.D.) on Vanuatu (17°S; 71°W) in the tropical South Pacific, and a previously unknown tropical volcano (1809 A.D.) that was first identified in both Greenland and Antarctica by *Dai et al.* [1991].

As more ice core-derived volcanic histories have become available, questions have been raised regarding the quantity and timing of material delivered to the polar ice sheets and how much reliance can be placed upon a reconstruction from one core. Knowledge of transport time and the relationship between the quantity of sulfur gases emitted and the SO_4^{2-} deposited is critical if the full potential of the ice cores for volcanic reconstruction and climate modeling constraints is to be realized. For example, a 4000-year volcanic history from the Plateau Remote (PR) core drilled high on the East Antarctic Plateau [*Mosley-Thompson,* 1996] raises a question about the relative magnitude of the sulfate flux from Tambora and Kuwae [*Cole-Dai et al.*, 2000, their figure 2]. At the PR site (84°S; 43°E; 3330 masl) the measured sulfate flux from Kuwae is five times that from Tambora, but the low annual average accumulation ($\sim$ 40 mm a^{-1} w.e.) is a complicating factor that calls for careful interpretation and more complementary histories. Undoubtedly, the PR record points to an event that may be very climatically

important, at least in the Southern Hemisphere if not globally, and it should be pursued more vigorously in other East Antarctic ice cores as well as in Greenland. Better dating and continuous analysis of multiple parameters in more cores will provide the robust volcanic sulfate flux histories necessary to place better constraints on the timing, duration and magnitude of stratospheric aerosol loads associated with specific eruptions.

3. THE GREENLAND PARCA CORES

A suite of spatially distributed cores collected since 1995 under NASA's Program for Arctic Regional Climate Assessment (PARCA) [*Thomas et al.*, 2001] now provides an unprecedented opportunity to assess local to regional variability of the various "climate" signals preserved within the Greenland ice sheet. PARCA cores are particularly valuable for their broad spatial distribution and the accurate dating of annual layers using multiple seasonally varying indicators ($\delta^{18}O$, insoluble dust, H_2O_2, nitrate, calcium). Figure 1 illustrates the locations of six multi-century cores collected as part of PARCA, as well as the Summit area where three cores (Site T, GISP2 and GRIP) discussed in the paper were collected, and the location of two older core sites (Crête, Dye 3) also mentioned in the text.

As the PARCA cores were drilled and analyzed primarily to reconstruct accumulation histories for different sites on the Greenland ice sheet [*Mosley-Thompson et al.*, 2001], their extensive analyses and careful interpretation resulted in very precise dating and determination of the thickness (mass accumulation) of each annual layer. Figures 2 and 3 illustrate the precision with which ice cores may be dated using multiple seasonally varying parameters. Figure 2A shows the section of the Site T Core 2 containing both Tambora and Unknown. The core (drilled in 1989 within 5 km of the GISP2 site) was analyzed continuously (top to bottom) for major anions (2039 samples) as well as for $\delta^{18}O$ (2075 samples) and dust concentrations (2075 samples). Due to the low annual average accumulation at Site T (~220 mm a^{-1} w.e.) the seasonal variations in $\delta^{18}O$ are smoothed by vapor diffusion in the firn [*Johnsen*, 1977]. The difficulty of using only $\delta^{18}O$ for dating cores from regions of low annual average accumulation (< ~250 mm a^{-1} w.e.) is readily apparent. Recently, physically-based models have been developed to 'back-diffuse' isotopic records [*Johnsen et al.*, 2000], making them more useful for dating at greater depths and in regions of lower accumulation. Fortunately, the seasonal variations in NO_3^- provided an important complement to the seasonal dust concentrations and allowed very accurate dating. Figure 2B illustrates the contemporaneous section from a 120-m core drilled in 1996 at the GITS site (formerly Camp Century) in northwest Greenland (see Fig. 1). The GITS core was dated using seasonal variations in $\delta^{18}O$ (2586 samples) and dust (2586 samples) measured at Ohio State by these authors and seasonal variations in H_2O_2, nitrate, and calcium measured by colleagues M. Anklin, R. Bales and J. McConnell, all then at the U. of Arizona, using their continuous melter system. Finally, the dating of the upper ~50 years of both cores was confirmed by identification of beta radioactivity horizons associated with thermonuclear testing in the 1950s and 1960s. A full discussion of the methods used to date the cores is provided by *Mosley-Thompson et al.* [2001, their figures 2 and 3].

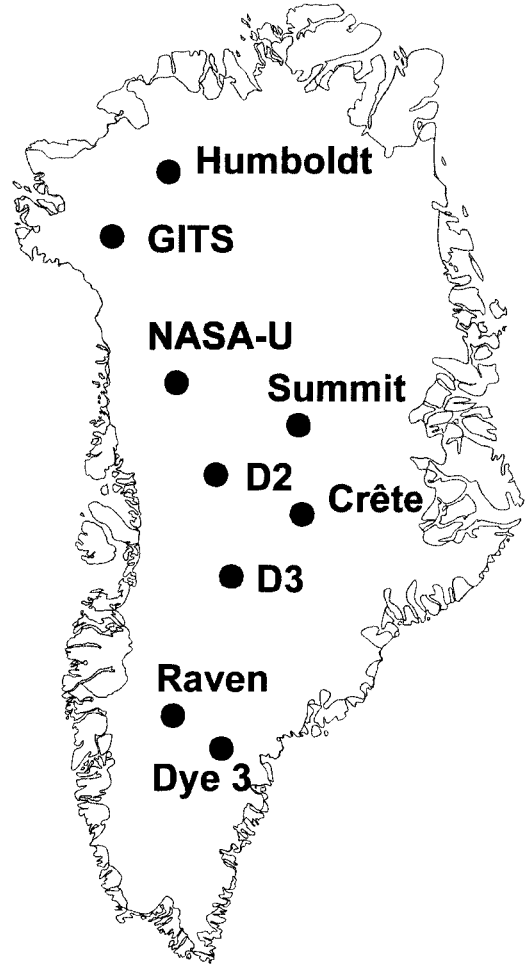

Figure 1. Shown are the locations of the six multi-century cores collected by the PARCA program, two older cores discussed in the text (Dye 3 and Crête) and the Summit Site where the GISP2, GRIP and Site T cores were drilled.

At some PARCA sites, multiple closely spaced cores demonstrate that signals of high-frequency (annual to possibly decadal scale) climate variability preserved in the ice sheet are partially masked by glaciological noise. Proxy records from the PARCA cores document that climate reconstructions, including inferences about volcanic emissions from a single core should be interpreted cautiously with application of appropriate filters to reduce local noise, and careful extrapolations from local to regional scales. The

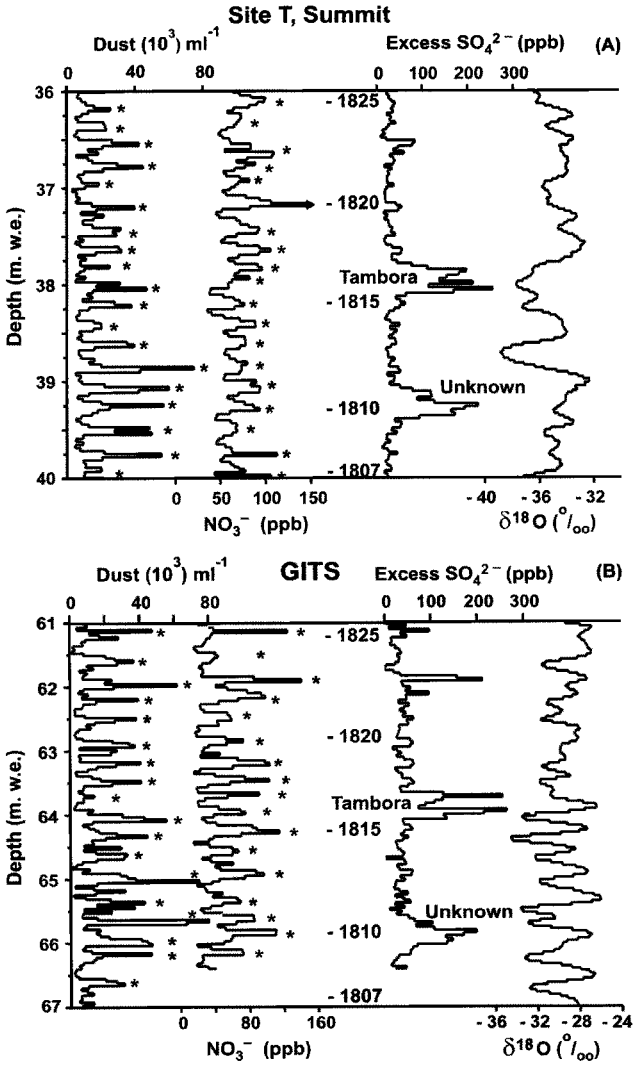

Figure 2. (A) Illustrated are the seasonal variations in dust, NO_3^-, and $\delta^{18}O$ used to date the entire Site T Core 2 and the SO_4^{2-} concentrations in the section of core containing the Tambora and Unknown eruptions; (B) The same four constituents are shown for the contemporaneous section of the GITS core that was dated using seasonal variations in dust, NO_3^-, $\delta^{18}O$, H_2O_2, and Ca^{2+}.

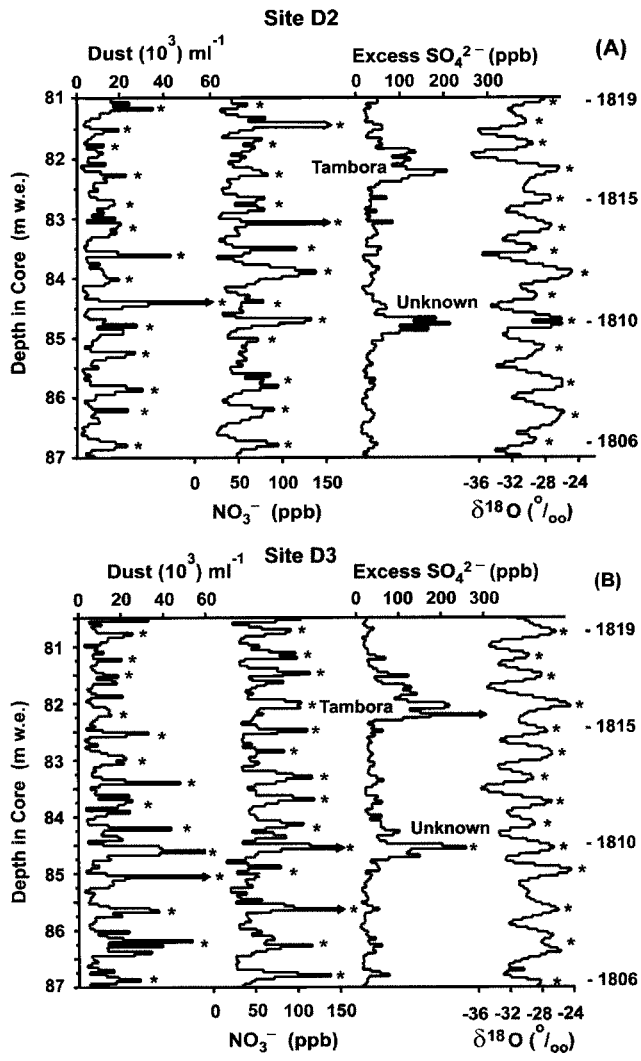

Figure 3. The seasonal variations in dust and $\delta^{18}O$ used to date both the D2 (A) and D3 (B) cores are shown along with the SO_4^{2-} and NO_3^- concentrations for the core sections containing the Tambora and Unknown eruptions.

data presented here represent our first results from an ongoing project to produce a complete (top to bottom), high resolution chemical characterization of five of the six multicentury PARCA cores, Humboldt, GITS, D3, D2 and Raven. These cores contain records extending back to 1153, 1717, 1745, 1740, and 1781 A.D., respectively. With the time scales firmly in hand, the SO_4^{2-} concentrations can be measured for sections of the cores containing known volcanic eruptions. Ultimately, the entire length of each core will be analyzed to isolate and identify all the volcanic horizons, including more modest eruptions, and to compare the timing and quantity of the volcanically derived sulfate

associated with individual events from site to site across Greenland.

3.1 Timing of the Unknown Pre-Tambora Eruption (1808 or1809?)

The PARCA cores allow the timing of the arrival of the volcanic sulfate associated with the unidentified pre-Tambora eruption (henceforth called Unknown) to be critically evaluated. The chemical analyses presented here were made by ion chromatographic analysis of samples that were cut from the core and then cleaned and melted under Class 100 clean room conditions. Details of the analytical method are described by *Dai et al.* [1995]. Based upon detailed chemical analysis of two cores, one from Siple Station in Antarctica and one from Site T in central Greenland, *Dai et al.* [1991] reported an explosive volcanic eruption in 1809 A.D. that does not appear in the historical records. As the eruption produced a sulfur rich plume that spread meridionally in the stratosphere to both polar regions the volcano is almost certainly located in the tropics. *Dai et al.* [1991] speculated that this event contributed to the decline in surface temperatures that preceded the eruption of Tambora in 1815 A.D. They concluded that the eruption occurred in 1809 A.D., one year prior to the 1810 A.D. arrival of the sulfate to both ice sheets, by assuming a one year delay in the arrival of sulfate at the poles similar to the delay observed for the emissions from the 1815 A.D. eruption of Tambora.

The timing of the ~1809 A.D. eruption has been questioned, primarily on the basis of the response of tropical marine air temperatures at this time [*Chenoweth*, 2001]. The only physical evidence for the (pre-Tambora) eruption comes from elevated sulfate layers preserved in both polar ice sheets [*Dai et al.*, 1991; *Clausen et al.*, 1997] and a climate response in both proxy and observed temperature records [*Briffa et al.*, 1994, 1998; *Chenoweth*, 2001, respectively]. *Briffa et al.* [1994] used maximum late wood density as a proxy for summer temperatures across northern North America. They reported an extremely cold summer in 1810 A.D. (second or third coldest depending upon the predictor used). In their Northern Hemisphere composite (383 chronologies in 8 regions) 1810 A.D. ranks as the 15th and 49th coldest summer in their NHD2 and NHD1 chronologies, respectively [*Briffa et al.*, 1998]. Recently, *Chenoweth* [2001] has suggested that the timing of the eruption is March to June of 1808 A.D., concurrent with a major cooling in two Malaysian observational land-based temperature records, and consistent with a maximum cooling of tropical marine air temperatures in 1808 A.D. Although the temperature response argument is intriguing, previously existing ice core data point to an 1810 A.D. arrival of the excess sulfate on both Greenland and Antarctica, supporting an eruption date in 1809 A.D.

A brief review of the Greenland ice core data reveals that *Clausen et al.* [1997] report acidity peaks at 1816 and 1810 A.D. in the central Greenland GRIP core. They did not find the latter event in their Dye 3 core, but *Langway et al.* [1995] reported elevated sulfate in 1810 A.D. in their two cores from the Dye 3 area. Five Greenland cores from the southern half of the ice sheet (Crête, Dye 3-81, Dye 3-18C, see *Langway et al.*, [1995]; GRIP, see *Clausen et al.*, [1997], and Site T, see Figure 2A) contain sulfate deposited in 1810 A.D. However, the chicken and egg issue raised previously cannot be discounted as volcanic layers were often used to guide and adjust initial time scales based on $\delta^{18}O$ and dust in earlier cores. The reader is encouraged to review the dating details for each core.

Figure 2 (A, B) confirms that the sulfate from the Unknown event arrives at these two sites (central and northwest Greenland) in 1810 A.D., consistent with earlier results from cores in central and southern Greenland. Comparing Site T and GITS it is also evident that the peak flux of sulfate in 1810 A.D. in both cores is comparable to that arriving in 1816 A.D., one year after the eruption of Tambora. The difference is that the Tambora sulfate remains much higher in the second year (1817 A.D.) while the sulfate concentrations in 1811 A.D. return nearly to pre-eruption levels. Although providing strong support for sulfate arrival in 1810 A.D., the GITS and Summit sites are not ideal for such high resolution inspection of the depositional record. The low annual average accumulation at Summit (~ 220 mm a^{-1} w.e.) increases the potential for the mixing of consecutive layers, although that does not appear to be a problem in the Site T Core 2. The GITS site is closer to the coast and although it is much further north, it is prone to modest surface melting in summer. In some sections of the GITS core, pipes (indicating past conduits for percolating water) and sills (areas of meltwater ponding and refeezing) were recorded. This has the potential to disturb the record although Figure 2B shows no sign of disturbance and the visible stratigraphy for that section of core was free of melt signals (e.g., clear, bubble-free ice).

Two of the PARCA cores, Site D2 (71°45'N; 46°20' W) and Site D3 (69°48'N; 44°00'W), are ideally situated in a region of higher accumulation that is also very cold and dry. These cores provide the best evidence to date for the arrival of Unknown's volcanic sulfate in 1810 A.D. In fact, these data should put the issue to rest. The D2 and D3 cores (drilled in 1999, see Fig. 1 for locations) were analyzed from top to bottom for $\delta^{18}O$ and dust. The D2 core was cut

into 2542 samples each for dust and $\delta^{18}O$ while the D3 core was cut into 3027 samples for each parameter. The time scales for both cores were constructed by counting the very distinct annual layers in dust and $\delta^{18}O$ and the dating of the upper 47 years (post-1953) was confirmed by identification of beta radioactivity horizons associated with thermonuclear testing. These sites have the advantage of high annual average accumulation (D2: 424 and D3: 488 mm a^{-1} w.e.) and lie well into the cold, dry snow facies (D2: 2640 masl; D3: 2560 masl) as evidenced by the absence of melt features. Figures 3A, B leave little doubt that the sulfate associated with Unknown was deposited in 1810 A.D. snowfall over Greenland and that most of the sulfate was deposited in a single year. We conclude that it is very unlikely that the eruption occurred in March-June 1808 as suggested by *Chenoweth* [2001].

It is important to mention that confirmation of the arrival time of Unknown's volcanic sulfate to Antarctica rests solely on evidence from two cores from sites with sufficiently high accumulation rates to decipher and firmly date an annual event. These are the previously mentioned Siple Station and Dyer Plateau cores [*Mosley-Thompson et al.*, 1991; *Mosley-Thompson*, 1992; *Thompson et al.*, 1994]. Although emissions from Unknown have been found in other Antarctic cores, the lower accumulation rates at those sites lead to dating imprecision and enhanced likelihood of mixing with adjacent snow layers. *Delmas et al.* [1992] report elevated sulfate in 1810 A.D. at South Pole where the annual average accumulation today is 85 mm a^{-1} w.e. [*Mosley-Thompson et al.*, 1999]. *Langway et al.* [1995] reported the same event as occurring in 1809 A.D. in their South Pole core and at 1811 A.D. in their 1989 core from Marie Byrd Land in West Antarctica (annual average accumulation reported as 100 mm a^{-1} w.e.). Clearly, low accumulation sites are not optimal for annual to sub-annual refinement of a depositional event.

3.2. Spatial Distribution of Excess Sulfate (EXS) Fluxes

Given the accurate dating of the PARCA cores discussed above, the annual flux of excess sulfate (EXS) to the ice sheet may be calculated for any volcanic event that has been chemically analyzed. As our preliminary analyses did not include cations (e.g., Na^+), the EXS reported here is calculated by subtracting the sea salt sulfate (SSS) determined by assuming that all of the Cl^- is derived from the ocean. This is obviously not the case and sodium (Na^+) is generally used for that calculation. The imprecision introduced by using Cl^- to calculate EXS depends upon the relative contribution of SSS to the total sulfate flux. Detailed chemical studies, including analyses of most cationic and anionic species, are available for only a few Greenland cores, most notably GISP2 and GRIP in central Greenland. *De Angelis et al.* [1997] report that the bulk of the sodium measured along the GRIP core is marine in origin suggesting that Na^+ would be preferable to Cl^- for estimating the marine contribution to the total SO_4^{2-}. Fortunately, use of Cl^- does not introduce a large difference in the calculated EXS concentrations because sea-salt sulfate comprises only ~5% of the total sulfate flux in central Greenland snow [*Legrand et al.*, 1997; *Saltzman et al.*, 1997]. This was verified by analyzing a 4-meter section of both the D2 and D3 cores for both anions and cations and calculating EXS using both Na^+ and Cl^-. The differences ($EXS_{Cl} - EXS_{Na}$) were 0.63 and –0.83 parts per billion (ppb) for D2 and D3, respectively and this is relative to their EXS_{Na} averages of 26.4 and 24.2 ppb, respectively. The negative sign results from the SSS contribution to EXS and the small values imply that using Cl^- rather than Na^+ makes virtually no difference (2.4% and 3.4% for D2 and D3, respectively). Thus, EXS concentrations reported here were calculated by subtracting from the total SO_4^{2-} the SSS obtained by multiplying the Cl^-concentration for each sample by the SO_2^- / Cl^- ratio in sea water (0.103). The remaining background concentration of EXS consists primarily of non-eruptive volcanic emissions and DMS emissions from marine biota [*Legrand et al.*, 1997]. The SO_4^{2-} derived from sulfur gases emitted by explosive volcanic eruptions will be superimposed as 'spikes' upon the more constant background EXS concentrations.

The annual EXS flux may be calculated by multiplying the EXS concentration in each sample by the sample's length in water equivalent and then adding all the sample fluxes for each year. The annual average background flux of non-volcanic EXS (Table 1) is calculated by averaging the non-volcanic years (excludes fluxes for 1817, 1816, 1811, and 1810 A.D. shown in Table 2). The time intervals for each background flux calculation are slightly different

Table 1. Average annual flux of background or 'non-volcanic' EXS and the average annual accumulation are shown for the time intervals indicated. Fluxes for 1817, 1816, 1811 and 1810 A.D. were removed.

Core	Time Interval	Annual Flux kg m^{-2}	Annual Accumulation mm w.e.
GITS	1825-1809	10.42	312
D2	1818-1806	12.06	424
D3	1819-1807	12.24	488
Site T	1822-1808	9.91	224
Raven	1819-1809	9.46	325

Table 2. Annual fluxes of excess sulfate in kg km^{-2} were determined at five Greenland sites for the eruptions of Tambora, Unknown and Laki. The calculations are described in the text.

Core	1817 kg m^{-2}	1816 kg m^{-2}	1811 kg m^{-2}	1810 kg m^{-2}	ratio 1810/ 1816	ratio 1811/ 1817	1784 kg m^{-2}	1783 kg m^{-2}
GITS	21.1	27.3	4.7	28.5	1.04	0.22	0.0	ua*
D2	21.4	30.9	6.0	32.0	1.04	0.28	0.0	222.0
D3	29.5	55.9	9.1	37.6	0.67	0.31	0.0	323.5
Site T	17.5	23.1	6.9	26.3	1.14	0.39	0.0	114.6
Raven	19.8	35.5	0.0	25.8	0.73	–	0.0	142.6
Average	21.9	34.5	6.7	30.0				
Humboldt	nm*	nm*	nm*	nm*			22.9	56.80
NASA-U	nm*	nm*	nm*	nm*			10.1	157.2

*nm means not measured and ua means unavailable. The section of the GITS core containing the Laki emissions was recovered in poor condition that made detailed (sub-annual) analysis impossible.

among the cores as only selected sections of the four PARCA cores have been chemically analyzed thus far. Few flux values are available for comparison as most Greenland sulfate fluxes have been calculated using either H^+ concentrations from ECM or acidity measurements and not SO_4^{2-} analyses. As previously mentioned ECM measurements provide rapid estimates of acidity that can be compared to evaluate the relative abundance of volcanic aerosols associated with different eruptions. However, as discussed above in section 2.1 and by *Zielinski* [1995] the ECM signal also reflects other acids such as HNO_3, HF, and HCL so that the volcanic sulfate determined by ion chromatographic (IC) analyses provides a much better basis for estimating stratospheric sulfate loading. Therefore, the comparisons below include only sulfate fluxes based on SO_4^{-2} concentrations from IC analyses.

Clausen et al. [1988] report background concentrations of H_2SO_4 (see their Table IV) associated with Laki (4 cores) that range from 6 to 11 kg km^{-2} and from 10 to 24 kg km^{-2} for Tambora at two sites. With one exception, 24 kg km^{-2}, their background fluxes (based on less than 3 years) are comparable to our SO_4^{-2} fluxes (range: 9.5 to 12.24 kg km^{-2}) based on 11 to 17 years (Table 1). Plotting the average background EXS concentrations (Table 1) versus the average w.e. accumulation (for the same time interval) at each of our sites yields a linear relationship ($R^2 = 0.753$, significance = 94%) strongly suggesting that SO_4^{-2} is deposited primarily by wet deposition (Fig. 4). A similar relationship was reported by *Legrand et al.* [1997, their figure 6] for both Antarctica and Greenland. Thus, the presence or absence of wet deposition at a site (i.e., whether it is snowing or not) contributes to the differences in EXS concentrations recorded from site to site for the same eruption.

The concentrations of background EXS for each core (Table 1) were subtracted from the annual EXS fluxes associated with 1817, 1816, 1811, and 1810 A.D. to determine the EXS derived from Tambora and Unknown. The resulting annual fluxes (Fig. 5 and Table 2) confirm that EXS in 1816 is fairly constant among the sites except for the much higher concentration at D3. Figure 3B shows one sample with very high SO_4^{2-} (344 ppb) in the Tambora horizon. The dispersion of the EXS flux among the sites is estimated by the ratio of the standard deviation (σ) to the mean (x) which is 0.33 and 0.15 for 1816 and 1810 A.D., respectively. The average EXS flux for all five cores is 34.5 kg km^{-2} a^{-1} in 1816 A.D. and 21.9 kg km^{-2} a^{-1} in 1817 A.D. The latter is ~64% of that deposited in 1816 A.D. By comparison, the average EXS flux in the second year following Unknown is much less, only ~22% of that in 1810 A.D. The average flux in 1810 (30 kg km^{-2} a^{-1}) was ~87%

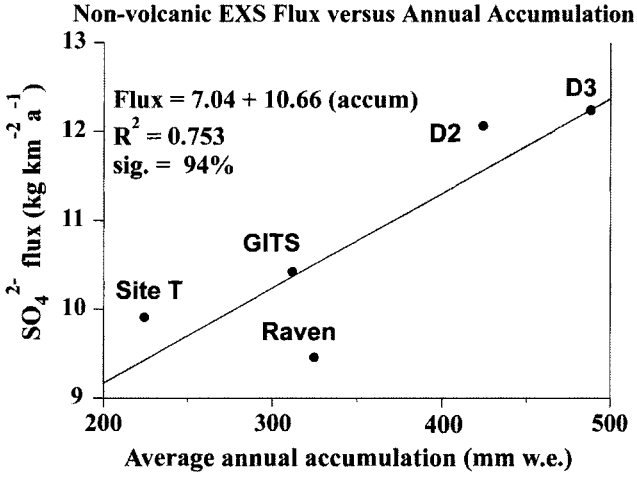

Figure 4. A strong linear relationship exists between the annual average background (non-volcanic) EXS concentrations and the average annual net accumulation. The time interval for calculating the averages from the five different ice cores is shown in Table 1.

of that deposited in 1816, confirming that the stratospheric sulfate burden in 1810 was large and undoubtedly affected hemispheric surface temperatures [*Briffa et al.*, 1994, 1998], if not global temperatures. However, the much reduced EXS in 1811 A.D. suggests that Unknown is unlikely to have had a strong direct effect on surface temperatures in the following year.

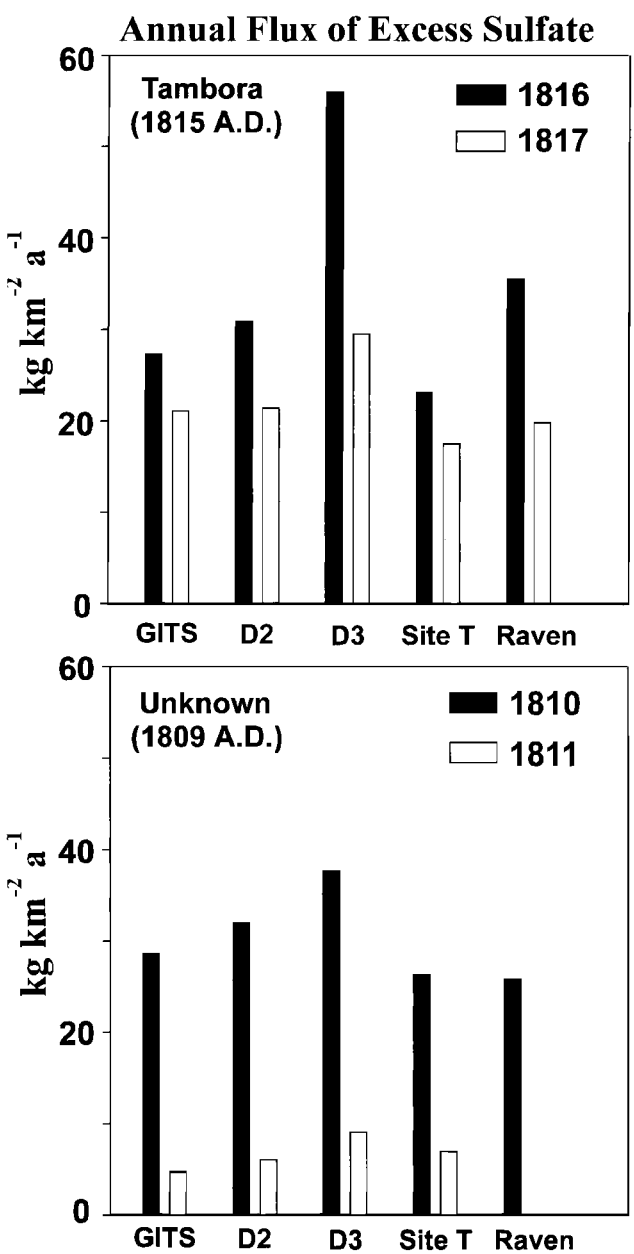

Figure 5. The annual fluxes of volcanically-derived EXS from Tambora and Unknown are compared for five Greenland ice core sites (locations shown in Fig. 1).

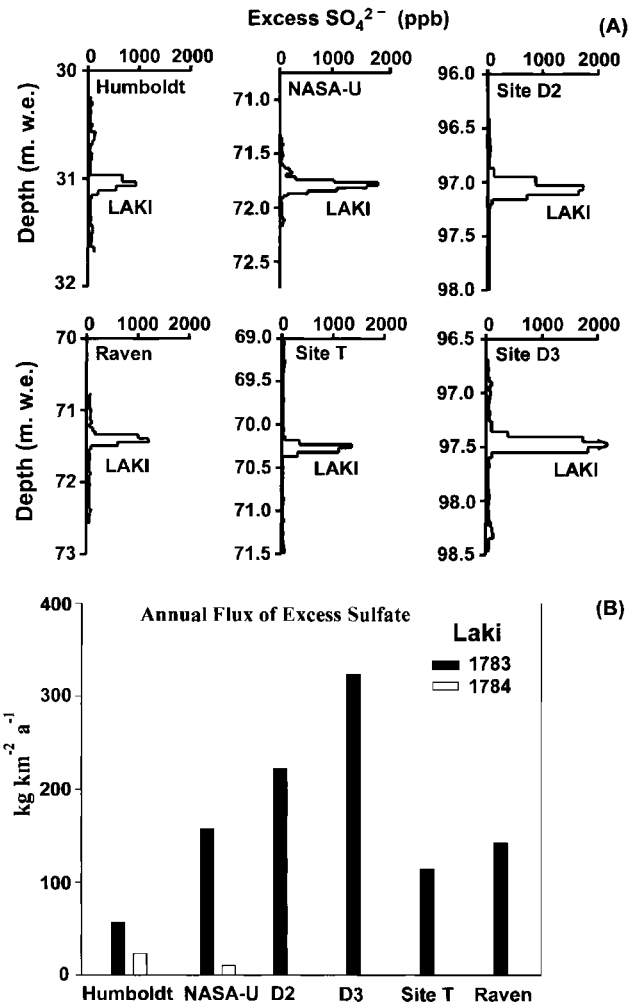

Figure 6. (A) The excess sulfate deposited at six Greenland ice core sites is shown before and after the eruption of Laki. (B) The annual flux of EXS deposited in 1783 and 1784 A.D. is compared for the six cores shown in (A).

The excess SO_4^{2-} and annual flux of EXS associated with the eruption of Laki (June 8, 1783 to February 7, 1784 A.D.) are shown in Figures 6A and 6B for four of the same cores as in Figure 5 plus the NASA-U and Humboldt cores (Fig. 1). The spatial distribution of the Laki fluxes (Fig. 6A,B) is more variable (σ/x = 0.5) than those for Tambora and Unknown (σ/x = 0.33 and 0.15, respectively). The SO_4^{2-} flux associated with Laki is similarly influenced by the rate of accumulation at the time of deposition (Fig. 7).

The EXS fluxes from Tambora and Laki can be compared for three different core sites (GRIP, GISP2 and Site T) in the Greenland Summit region. For the GRIP core *Clausen et al.* [1997] report volcanic acid deposition (principally sulfate)

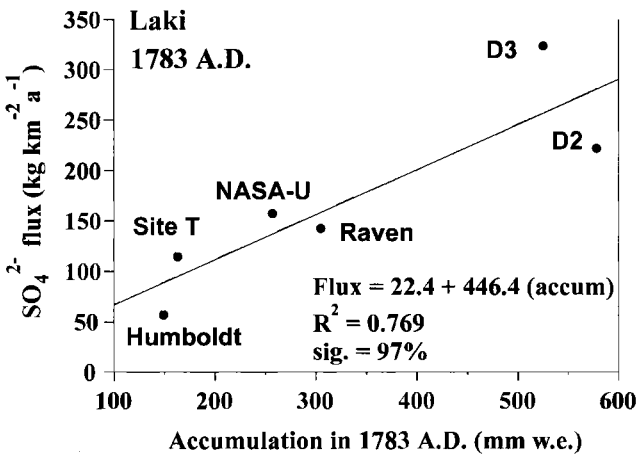

Figure 7. The strong linear dependence of the annual flux of EXS from the eruption of Laki and the annual average accumulation in that year is illustrated for six Greenland ice core sites (Fig. 1).

of 35, 25 and 147 kg km^{-2} for Tambora, Unknown and Laki, respectively. The comparable fluxes for Site T (Table 2) are 23.1 (1816), 17.5 (1817), 26.3 (1810), 6.9 (1811), and 114.6 kg m^{-2} (1783). These compare well although it is not clear whether their Tambora and Unknown fluxes are for just the first year post-eruption, or for both years post-eruption. A similar comparison is made with the *Zielinksi* [1995] fluxes from the GISP2 core from which he reports a total volcanic SO_4^{2-} flux (background removed) of 36 kg km^{-2} for the two year event. This compares well with our Site T total flux of 40.6 kg km^{-2} (Table 2, sum of 1816 and 1817 A.D.) for the Tambora eruption. Likewise, the GISP2 flux for Unknown compares well with the Site T flux (28 kg km^{-2} versus our 33.2 kg km^{-2}). The sulfate fluxes associated with Laki are much more variable among the three Summit cores. The GISP2 flux of 57 kg km^{-2} is significantly less than 114.6 kg km^{-2} deposited at Site T and both of these are less than 147 kg km^{-2} deposited at the GRIP site ~30 km east. The depositional differences in Laki sulfate could reflect a more variable (less well mixed) aerosol mass transported directly to the ice sheet via the troposphere over the 7 month duration of the eruption. As wet deposition is the primary mechanism for removal from the atmosphere, whether it is snowing at one site versus another as the aerosol mass transits the site could also affect the spatial differences. Although other explanations could be proposed it is likely that the overriding factor is the proximity of Laki to Greenland, versus the tropical locations of Tambora and Unknown that result in a much better mixed stratospheric volcanic aerosol mass.

The timing of the arrival of Laki sulfate has been assumed to be 1783 A.D., but *Fiocco et al.* [1994] have questioned this based upon their identification of glass shards from Laki that were found just below (preceding) the sulfate horizon in the GISP2 core (Summit). They suggest that the sulfate arrived in the summer and early fall of 1784, one year after the eruption. Since their result is based upon evidence from a single site where the low annual average accumulation (~220 mm a^{-1} w.e.) allows greater mixing of consecutive layers, their results must be viewed cautiously until confirmed in multiple cores at sites with higher accumulation rates.

The acidity layer associated with Laki has served as a cornerstone for nearly all Greenland ice core chronologies and has been routinely assigned to 1783 A.D.; therefore, the timing of the sulfate deposition must be independently verified as it has been for the Unknown eruption. This requires the analyses of multiple seasonally varying parameters, along with high resolution continuous anion and cation chemistry, over the entire section from 1820 A.D. down to 1780 A.D. in cores with higher accumulation and no melt disturbance. Only by such a methodical examination will the timing of the sulfate from Laki be firmly resolved. This has important implications for reconstructing the atmospheric transport pathway and sulfate distribution over the Northern Hemisphere following the eruption of Laki. The cores from PARCA sites D2 and D3 are ideal for testing the arrival of Laki sulfate and the existence of Laki ash and this investigation is now underway at Ohio State.

4. CONCLUSIONS

The anion analyses of the multi-century PARCA cores complement previous results that were more spatially restricted. These new data confirm that the sulfate aerosols from the unidentified pre-Tambora eruption called Unknown (1) were widely dispersed across the Greenland ice sheet, (2) first arrived in the 1810 A.D. snow fall, and (3) in the first year post-eruption (1810 A.D.) produced excess SO_4^{2-} concentrations comparable to those produced by Tambora in the first year post-eruption (1816 A.D.), but much less EXS in the second year post-eruption (1811 A.D.). There are more modest eruptions yet to be identified and still more for which the excess sulfate flux and its arrival time over Greenland need to be established. The annual resolution of multiple seasonally varying parameters in the PARCA multi-century cores will help resolve timing issues and their broad spatial distribution will provide a more spatially coherent estimate of the EXS flux associated with specific eruptions. These EXS estimates underpin calculations of the atmosphere's volcanic aerosol load that is a required input for modeling the climate system's range of responses to volcanic forcing. The PARCA multi-century cores will expand and refine the Northern

Hemisphere volcanic history and as similar records of equivalent temporal resolution from Antarctica become available, their combination will provide a richer, more detailed, and thus more climatologically valuable global history of explosive volcanism.

Acknowledgments. The PARCA cores from Humboldt, GITS and Site T were drilled by personnel from the University of Nebraska Polar Ice Coring Office (PICO) who also provided the logistical support in all years under contract to NASA. The PARCA cores at Raven, D2 and D3 were collected using The Ohio State electro-mechanical drill designed and constructed by Victor Zagorodnov. We thank M. Anklin, R. Bales, J. Box, J. Burkhart, M. Davis, R. Edwards, K. Henderson, J. Kyne, Z. Li, P.-N. Lin, S. Lin, J. McConnell, A. Nolin, B. Snider, K. Steffen, and V. Zagorodnov for their contributions to the field and analytical aspects of the project. The quality of this paper was enhanced by discussions with Greg Zielinski, suggestions made by two anonymous reviewers and detailed and constructive comments offered by Caspar Ammann. The OSU contributions to PARCA (1996 - 2001) were supported by NASA grants NAG5-5032 and 6817 to The Ohio State University. This is contribution 1270 of the Byrd Polar Research Center.

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