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Synchronous deposition of volcanic ash and sulfate aerosols over Greenland in 1783 from the Laki eruption (Iceland)

Lijia Wei,[1,2] Ellen Mosley-Thompson,[1,2] Paolo Gabrielli,[1,3] Lonnie G. Thompson,[1,3] and Carlo Barbante[4,5]

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[1] Sulfate aerosols from the 1783–1784 A.D. Laki eruption are widely used as a reference horizon for constraining Greenland ice core time scales, yet the timing of the arrival of the sulfate remains under discussion. Two ice cores from western Greenland, analyzed with high temporal resolution, confirm that sulfate aerosols arrived over Greenland late in 1783, concomitant with the tephra, elevated concentrations of Cd, Bi, and Tl, all indicators of volcanic emissions, and with a short-lived Rare Earth Elements anomaly. Thereafter sulfate deposition declined rapidly. Very modest concentrations of sulfate in 1784 snowfall, evident in six Greenland cores, suggest a relatively short (less than 1 year) atmospheric residence time and an injection height limited to the lower stratosphere. An improved estimate of the associated stratospheric sulfate burden is calculated and provides an important input for models assessing climatic impacts of this volcanic eruption.
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1. Introduction

[2] The Greenland Ice Sheet (GIS) preserves an exceptional record of Earth's paleoclimatic and paleo-environmental conditions covering more than one hundred thousand years. The average annual accumulation rate (A_n) for the GIS is ~300 mm water equivalent (w.e.) a^{-1} [*Bales et al.*, 2001], but varies from very low values (<100 mm w.e. a^{-1}) in the northeast, to modest values (~230 mm w.e. a^{-1}) in central Greenland to higher values (>350 mm w.e. a^{-1}) in the south and regions close to the coast. In regions of low accumulation, surface and post-depositional processes are more likely to distort preserved seasonal indicators (e.g., stable isotopic ratios and H_2O_2) [*Johnsen*, 1977; *Neftel*, 1996; *Wolff*, 1996], and/or remove all or part of a year's accumulation, leading to unavoidable dating errors and uncertainty. Higher accumulation allows paleo-records to be better preserved and reconstructed more accurately, often with annual resolution. Cross-dating with multiple seasonally varying parameters also contributes to a more robust time scale. To clearly decipher proxy records so that they may be calibrated using meteorological data and then subsequently used to extend our knowledge of climatological processes further into the past, a tightly constrained time scale is essential. For example, reconstructing high frequency events, such as the North Atlantic Oscillation, El Niño, and La Niña, requires very accurate dating. Auxiliary material[1].

[3] Large explosive volcanic eruptions often result in the deposition of sulfate aerosols across the polar ice sheets. Some known volcanic horizons, such as those associated with the 1783–1784 Laki eruption and the 1815 Tambora eruption [*Cole-Dai et al.*, 2000], have been used for decades to constrain the time scales for Greenland ice cores [*Hammer et al.*, 1981; *Clausen and Hammer*, 1988; *Laj et al.*, 1990]. *Fiacco et al.* [1994] reported that ash particles from Laki arrived in 1783, while the associated sulfate pulse arrived in 1784 A.D. Their evidence consisted of a few glass shards found just below the layer with elevated sulfate concentrations in the GISP2 core from central Greenland (A_n = ~220 mm w.e. a^{-1}). Their dating of the core section containing this event was based solely on three Cl^-/Na^+ peaks interpreted as annual (1783–1785). Their inferred 1784 snowfall contained an almost negligible increase in dust and lacked the characteristic summer $\delta^{18}O$ peak. Although their evidence for the 1784 annual layer was ambiguous, their conclusion has been used to assess the 1783–1784 atmospheric sulfate loading, with ~12.5% of the total H_2SO_4 aerosols estimated to have been retained in the lower stratosphere for more than a year [*Thordarson and Self*, 2003]. A recent model simulation of the sulfate aerosol distribution by *Oman et al.* [2006] suggested that the majority of Laki sulfate arrived over the GIS in 1783, not in 1784. *Oman et al.* called for re-examination of the *Fiacco et al.* data and/or data from other ice cores to resolve the potentially spurious Cl^-/Na^+ peak (1784) that they suggested may reflect the emission of volcanic Cl^-. Two ice cores from high accumulation regions have been continuously analyzed and dated using multiple seasonally varying constituents [*Mosley-Thompson et al.*, 2001], and thereby provide an optimal data set to explore the arrival of Laki emissions over Greenland.

2. Data and Methods

[4] The two cores that underpin this investigation, D2 and D3, were collected in 1999 on the west side of the ice

[1]Byrd Polar Research Center, Ohio State University, Columbus, Ohio, USA.
[2]Atmospheric Sciences Program, Department of Geography, Ohio State University, Columbus, Ohio, USA.
[3]School of Earth Sciences, Ohio State University, Columbus, Ohio, USA.
[4]Institute for the Dynamics of Environmental Processes-CNR, University Ca'Foscari of Venice, Venice, Italy.
[5]Department of Environmental Sciences, University Ca'Foscari of Venice, Venice, Italy.

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[1]Auxiliary materials are available in the HTML. doi:10.1029/2008GL035117.

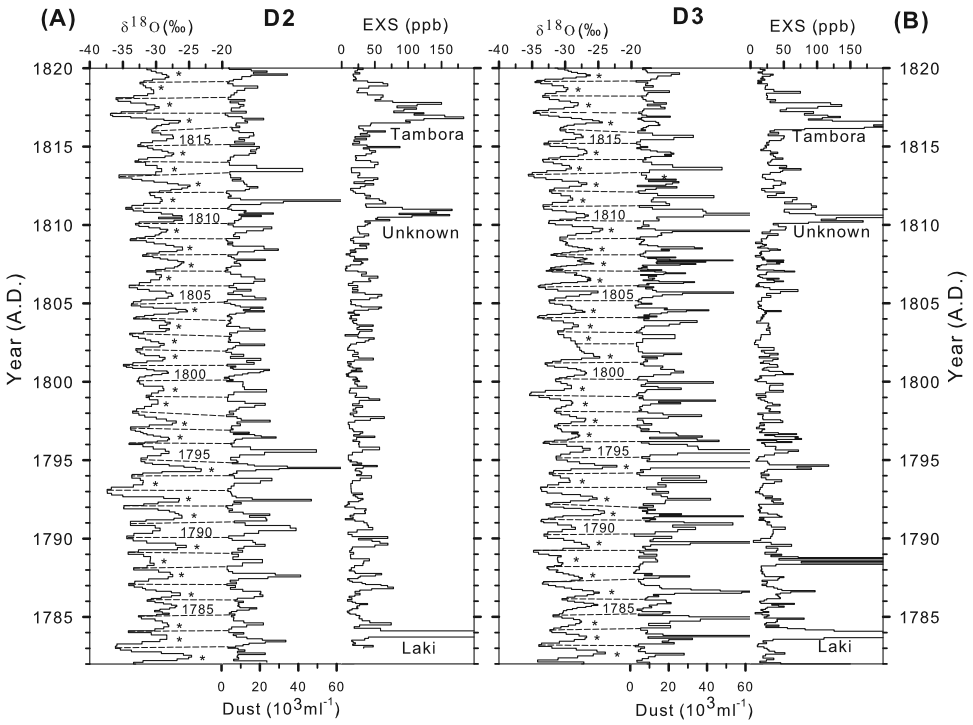

Figure 1. Seasonal variations in $\delta^{18}O$ and insoluble dust used to date the entire core are shown along with the excess sulfate (EXS) for the time interval 1782–1819 from the (a) D2 and (b) D3 ice cores.

divide, as part of the PARCA Project [*Thomas*, 2001]. Their A_n rates (1781–1999 A.D.) are 449 mm w.e. a^{-1} (D2) and 451 mm w.e. a^{-1} (D3). For location and details about these cores and the four additional cores used later for the Laki flux calculations, see *Mosley-Thompson et al.* [1993, 2001]. The oxygen isotopic ratios ($\delta^{18}O$), insoluble dust content, and major ions (Figure 1) were analyzed by the Ice Core Paleoclimate Research Group at The Ohio State University's Byrd Polar Research Center. A Finnigan-Mat Delta Plus mass spectrometer provided the $\delta^{18}O$ while the insoluble dust and major ion concentrations were analyzed with a Coulter Mulitsizer and Dionex ion chromatographs, respectively. This equipment is housed in a Class 100 clean room. Standard ion chromatography procedures [*Cole-Dai et al.*, 2000] were employed. Sections of the D3 core deposited one year before, during, and two years after the Laki eruption (1782–1785) were examined for tephra. These 16 samples, 4 per year (Figure 2), were cut, melted, and filtered onto 0.2 μm Nuclepore membranes for examination with Quanta and XL-30 ESEM scanning electron microscopes. Thirty-two samples were extracted from the D2 core and prepared in the Class 100 clean room using established ultra-clean procedures for trace element analyses [*Gaspari et al.*, 2006]. The analysis of Cd, Bi, Tl, and Rare Earth Elements (REE) were conducted by Inductively Coupled Plasma Sector Field Mass Spectrometry (Element2, Thermo) at the University of Venice [*Gabrielli et al.*, 2006]. The concentrations of excess Cd, Bi, and Tl were calculated by correcting for the terrestrial and marine contribution using the concentrations of Ba and Na, respectively.

[5] Generally, volcanic events are identified by elevated excess sulfate (EXS) deposited when gaseous compounds from explosive volcanic eruptions are oxidized to sulfuric acid [*Mosley-Thompson et al.*, 2003]. The EXS is routinely calculated by subtracting the sea-salt sulfate, estimated using either chloride or sodium, from the total SO_4^{2-}. The annual flux of EXS is calculated as $\sum_{i\in\{1\ year\}} c_i l_i$ for each core, where c_i = the concentration of EXS in each sample and l_i is the length of each sample in w.e. To remove the background sulfate flux, the non-volcanic sulfate flux was calculated by averaging the 5 years before the eruption (1778–1782 A.D.) and 5 years after 1784 (1785–1789 A.D.). We opted to use this smaller interval for the background calculation rather than the entire core to avoid more recent anthropogenic contributions to sulfate. The sulfate flux associated with the Laki eruption was then calculated as the difference between the total flux (1783 and 1784) and the calculated background concentrations (Table 1).

3. Results and Discussion

[6] Although the Laki eruption lasted 8 months (June 8, 1783 to Feb. 7, 1784), ~96% of the SO_2 was released during the first 5 months [*Thordarson and Self*, 2003], producing substantial societal and environmental impacts over Europe, North America, and the Arctic. Located on the west-central side of Greenland, the D2 and D3 sites receive annual accumulation twice that in the Summit region (GRIP, GISP2 cores), which greatly enhances the likelihood of complete preservation of the seasonally varying constituents. The timing of the arrival of Laki emissions was independently established using multiple seasonally varying parameters as illustrated for sections of cores D2 and D3 from 1782 to 1820 A.D (Figure 1). The dating was based primarily on seasonal variations in $\delta^{18}O$ and dust content. NO_3^-, another seasonally varying constituent (not shown), was also used to assist with the dating. Generally,

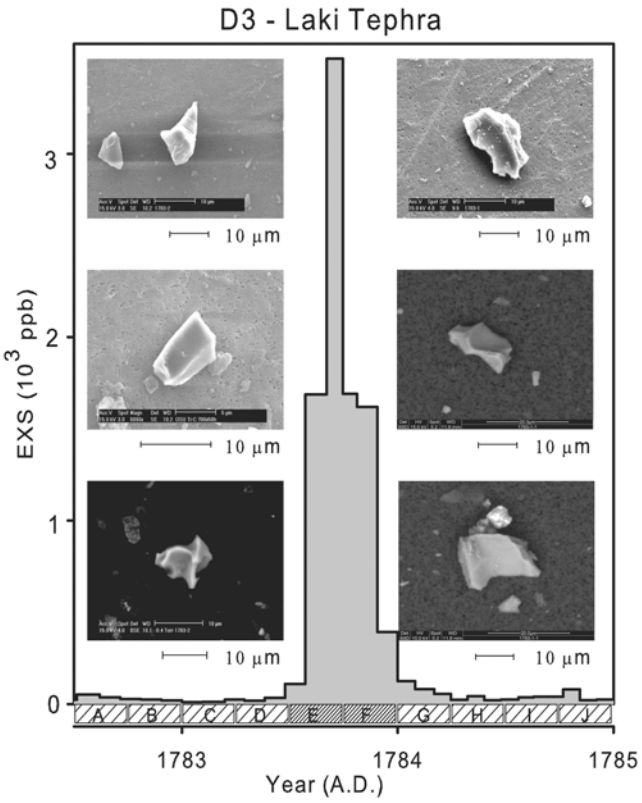

Figure 2. SEM photographs document the Laki tephra found in only two of the 16 filtered samples cut from the D3 core (shaded boxes on the time axis). Photographs E-1 to E-3 and F-1 to F-3 are from filters E and F (in fine shading), respectively. These two filters with tephra contain snow deposited in the second half of 1783, coincident with the EXS peak (also shown). No volcanic fragments were found on the remaining 14 filters.

$\delta^{18}O$ is lowest (most negative) in late winter (~February) and highest (most enriched or less negative) in mid to late summer (July to August) [*Bolzan and Strobel*, 1994; *van der Veen and Bolzan*, 1999]. Dust content usually peaks in spring and is lowest in winter [*Hammer*, 1977]. Both parameters were well preserved in D2 and D3 with little smoothing and disturbance. The top of this section is tightly constrained by the well dated 1816 EXS horizon from the 1815 eruption of Tambora [*Clausen and Hammer*, 1988; *Cole-Dai et al.*, 2000]. In both polar regions, the EXS peaks associated with Tambora arrived in 2 sequential annual pulses, 1816 and 1817 A.D., due to the height of injection into the tropical stratosphere and their long-distance transport poleward. Another large explosive tropical eruption, called the Unknown (1809) event, also provides an excellent time constraint. EXS from 'Unknown' arrived in both Antarctica and the Arctic in the 1810 A.D. snow fall [*Cole-Dai et al.*, 2000], producing EXS concentrations comparable to those deposited in 1816 A.D from Tambora [*Mosley-Thompson et al.*, 2003]. By using these seasonal indicators and reference horizons, Laki sulfate is determined to have arrived over Greenland late in 1783.

[7] In addition to the arrival of the EXS (Figure 1), the timing of tephra deposition in Greenland relative to the sulfate peak has implications for atmospheric transport processes. In D3, tephra fragments were found only in the layer coincident with the bulk (~97 to 98%) of the associated Laki sulfate peak, corresponding to the latter half year of 1783 A.D. (Figure 2 and Table 1). No fragments with distinctive volcanic features were observed on the remaining 14 filtered samples. Note that both D2 and D3 contain a prominent $\delta^{18}O$ summer peak (Figure 1) inconsistent with the assumption of strong summer cooling invoked by *Fiacco et al.* [1994] to explain the missing $\delta^{18}O$ summer peak in their core. In D2 and D3, there is a distinct two-sample peak in Cl^- (Figure 3d) that creates a spurious Cl^-/Na^+ signal late in 1783 (Figure 3e). In general, volcanically derived Cl^- is quickly scavenged by wet deposition before reaching the stratosphere [*Tabazadeh and Turco*, 1993]. However, due to their close proximity it is quite likely that some of the Cl^- emitted from Laki did arrive over the GIS creating the Cl^-/Na^+ peak erroneously identified as annual by *Fiacco et al.* [1994, Figure 4]. In fact, as shown in Figure 3, Cl^-/Na^+ is not a reliable indicator of seasonality and should be avoided as a dating tool for Greenland ice cores.

[8] The Laki eruption also contributed additional trace elements to the atmosphere. *Hong et al.* [1996] reported that excess Cd, Cu, and Zn were detected in the Laki fallout layer in the Greenland Summit core (A_n = ~230 mm w.e. a^{-1}) and that their deposition was constrained to a brief period. Their maximum Cd concentration was 12.6 ppt, coincident with the core section with a high electrical conductivity (an indicator of acidity). In our D2 core, the first peak of excess Cd (10.4 ppt) is coincident with the 1783 dust peak, and excess Cd reaches its maximum (80 ppt) near the end of 1783 (Figure 3). Other important volcanic tracers, such as excess Bi and Tl [*Matsumoto and Hinkley*, 2001], peaked at 9.3 ppt and 4.6 ppt, respectively, near the end of 1783 and quickly returned to their background values in 1784. Bi is derived mainly from volcanic emissions to the atmosphere and its average background concentration in

Table 1. Estimated Background EXS and Laki Eruption EXS in 6 Greenland Ice Cores

	A_n (mm w.e. a^{-1})			Bkg. EXS Flux[b]	Annual EXS Flux (kg/km^2)			Laki EXS Flux (kg/km^2)		EXS Flux[c] (%)
Cores[a]	(1781–1985)	1783	1784	(kg/km^2)	1782	1783	1784	1783	1784	1784
D3	451	525	323	18.9	11.5	314.4	24.6	295.5	5.7	1.9
D2	449	596	442	12.7	10.3	249.1	16.2	236.4	3.5	1.5
NASA-U	344	208	312	11.0	11.0	167.0	20.6	156.0	9.6	5.8
Dye 2	316	304	304	9.5	7.6	154.7	14.7	125.2	5.2	4.0
Site A	228	244	161	4.8	7.3	176.0	3.1	171.2	–	–
Site T	221	261	260	7.5	7.7	123.0	9.2	115.5	1.6	1.4

[a]Locations are shown by *Mosley-Thompson et al.* [2001, Figure 1].
[b]Background EXS flux is the average from 1778 to 1789, excluding 1783 and 1784.
[c]Percent of EXS flux in 1784, relative to the total flux for 1783 and 1784.

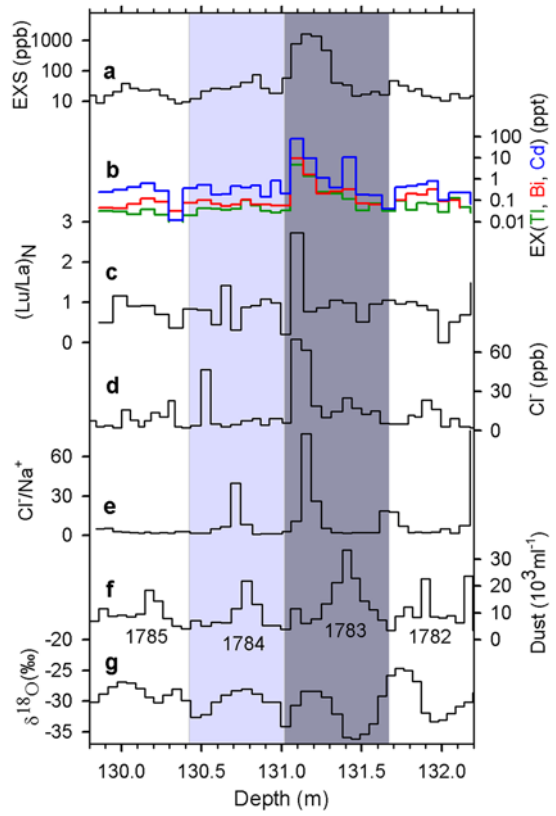

Figure 3. Section of Core D2 (1782 to 1785 A.D.) containing the Laki eruption: (a) calculated concentrations of excess sulfate (EXS); (b) calculated concentrations of excess Tl (gray solid line), Bi (dotted line), and Cd (dark solid line); (c) normalized Lu/La ratio; (d) measured concentration of Cl^-; (e) calculated ratio of Cl^- to Na^+; and (f) and (g) seasonal variations in the insoluble dust concentrations and $\delta^{18}O$, respectively, used to construct the time scale. The shading highlights 1783 (darker) and 1784 (lighter).

central Greenland snow is 0.15 ppt [*Ferrari et al.*, 2000], consistent with the Bi background concentration in D2 (0.03 to 0.34 ppt in 1782, 1784–1785). The background concentration of Tl in D2 is 0.01 to 0.13 ppt (also in 1782, 1784–1785), consistent with the Tl background (0.03 to 1.3 ppt) from Devon Island (Arctic) ice samples [*Krachler et al.*, 2005]. Generally, Cd, Bi, and Tl have low mean element weight ash fractions (0.1–3%) [*Aiuppa et al.*, 2003], indicating that these trace metals were transported to the GIS primarily by aerosols. In contrast, REE are refractory and usually associated with solid particles in precipitation [*Freydier et al.*, 1998], consistent with the behavior of the crustal normalized Lu/La ratio that reflects a change in dust composition. A brief peak of $(Lu/La)_N$, calculated as $(Lu/La)_N = (Lu_{sample}/Lu_{crust})/(La_{sample}/La_{crust})$ [*Wedepohl*, 1995], is observed late in 1783, in close correspondence with other elevated trace elements (Figure 3). Thus, the contemporaneity of tephra and the rapid change of the $(Lu/La)_N$ ratio strongly suggest that the Laki ash arrived over the GIS late in 1783, along with the elevated concentrations of Cd, Bi, Tl, and sulfate aerosols. These ice core-derived observations agree well with the model simulation by *Oman et al.* [2006] and confirm the rapid transport of volcanic emissions to Greenland.

[9] The decay time of volcanogenic sulfate signals in ice cores may provide information regarding the injection height of SO_2 gas by the eruption and the residence time of sulfate aerosols in the upper troposphere/lower stratosphere. The EXS associated with Laki in both 1783 and 1784 was calculated along with the background concentration of EXS for six Greenland ice cores (Table 1). The percent of the total EXS flux (1783 and 1784) from Laki arriving over Greenland in 1784 ranges from near zero to 6%, consistent with the portion of SO_2 estimated to have been produced by Laki during the last 3 months of the eruption (~4% from Nov. 1783 to Feb. 1784). *Thordarson and Self* [2003] estimated that 200 Mt of sulfate aerosols were produced by Laki and that ~25 Mt (12.5%) stayed aloft near the tropopause for >1 year. However, the relatively short atmospheric lifetime of the sulfate aerosols calculated from these 6 ice cores (Table 1) indicates that a much smaller portion of Laki aerosols remained in the lower stratosphere in 1784, suggesting both a relatively low injection height restricted to the lower stratosphere, and a short residence time (<6 months) for most of the emissions from Laki.

4. Conclusions

[10] Two well-dated Greenland ice cores from high accumulation regions confirm that most of the excess sulfate associated with the 1783 Laki eruption was transported quickly (in less than 6 months) to the GIS. Laki tephra fragments were found only in the annual layer containing the EXS peak. Volcanically-derived trace elements, Cd, Bi, and Tl, were elevated well above their natural background concentrations in snowfall arriving late in 1783, coincident with the main pulse of EXS. A brief $(Lu/La)_N$ peak was also observed late in 1783, indicating the changes in dust composition. These data virtually confirm that the volcanically derived ash particles, sulfate aerosols, and trace elements from Laki arrived almost simultaneously over Greenland late in 1783. The rapid decline in EXS in 1784, confirmed in six Greenland cores, suggests the injection height was limited to the upper troposphere/lower stratosphere and that most of the volcanically derived debris was removed within 6 months of the initial eruption.

[11] **Acknowledgments.** We thank Victor Zagorodnov, who was in charge of drilling of the D2 and D3 cores, Zhongqin Li, who conducted the dust analyses, and P. N. Lin, who conducted the $\delta^{18}O$ analyses. The D2, D3 and NASA-U cores were collected by the NSF-NASA PARCA Project, and the dust and $\delta^{18}O$ analyses were supported by NASA-NAG-5072 and -6817. NSF-OPP-0352527 provided graduate student support for Lijia Wei, who conducted the chemical analyses of D2 and D3. Trace element analyses were supported by the Consorzio per l'Attuazione del Programma Nazionale delle Ricerche in Antartide in Italy, under projects on Environmental Contamination and Glaciology. This is Contribution 1371 of the Byrd Polar Research Center.

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C. Barbante, Institute for the Dynamics of Environmental Processes-CNR, University Ca'Foscari of Venice, I-30123 Venice, Italy.

P. Gabrielli, E. Mosley-Thompson, L. G. Thompson, and L. Wei, Byrd Polar Research Center, Ohio State University, Columbus, OH 43210, USA. (wei.103@osu.edu)