

RESEARCH LETTER

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Key Points:

- Tephra from Changbaishan Millennium eruption is present in Greenland ice
- Associated sulfate levels suggest eruption was not climatically effective

Supporting Information:

- Introduction: Tephra geochemical and ice core chemical analyses reported in this paper.
- Readme_final

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Ash from Changbaishan Millennium eruption recorded in Greenland ice: Implications for determining the eruption's timing and impact

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Abstract Major volcanic eruptions can impact on global climate by injecting large quantities of aerosols and ash into the atmosphere that alter the radiative balance and chemical equilibrium of the stratosphere. The Millennium eruption of Tianchi (Paektu), China/North Korea, was one of the largest Late Holocene eruptions. Uncertainty about the precise timing of the eruption has hindered the recognition of its climate impact in palaeoclimate and historical records. Here we report the compelling identification of the eruption's volcanic signal in Greenland ice cores through the association of geochemically characterized volcanic glass, represented in by bimodal populations that compare with proximal material from the source eruption. The eruption most probably occurred in the A.D. 940s, 7 years after the Eldgjá eruption on Iceland. We examine the eruption's potential for climate forcing using the sulfate records from the ice cores and conclude that it was unlikely to have had a global or extraregional impact.

1. Introduction

Volcanic forcing of climate at subdecadal scales, mainly through stratospheric loading of sulfates and other aerosols and their impact on radiative balance, is well established through instrumental records of recent eruptions [McCormick *et al.*, 1995; Fischer *et al.*, 2007], although the precise mechanisms by which this is achieved remain uncertain [Driscoll *et al.*, 2012]. Understanding the nature and causes of past climate change nevertheless requires that climate models factor in atmospheric loading of aerosols [Schmidt *et al.*, 2012; Timmerck, 2012]. Polar ice cores provide the most important records of past volcanism for paleoclimate models through their acidity levels, chemistry and electrical conductivity, enabling stratospheric sulfate loading to be estimated [Robock and Free, 1995; Gao *et al.*, 2008]. Neither the location nor the magnitude of a source eruption can be determined from ice core chemical records alone, however, as less climatically effective high-latitude eruptions can also contribute to the ice core acidity levels [Robock and Free, 1995]. Ash (tephra) particles in the ice, conversely, can provide evidence of the source eruption and greatly improve the interpretation of likely volcanic forcing.

Changbaishan Volcano (also known as Tianchi, Baitoushan, or Paektu) (128°03'E, 42°00'N) is an intraplate stratovolcano located on the border between China and North Korea. A Plinian eruption around 1000 years ago (the "Millennium eruption") has been attributed a volcanic explosivity index of 7 based on an estimated ~25–35 km high eruptive column and a total tephra volume of ~100 km³ (magma volume (DRE ~25 km³)) [Horn and Schmincke, 2000; Wei *et al.*, 2003]. Fine fallout from this eruption extended mainly to the east and east-northeast, as far as north Japan and Kuril trench (>1000 km), where a 2–10 cm thick deposit of ash has been recorded [Machida and Arai, 1983] (Figure 1). Modeling the radiative forcing of the eruption, Li *et al.* [1996] estimated an average Northern Hemisphere cooling of 0.85 °C and a potential global climate impact extending over 6 years. Notwithstanding uncertainty regarding the eruption's atmospheric loading, estimates of halogen emissions based on petrological data indicate potential for stratospheric ozone depletion [Horn and Schmincke, 2000]; although sulfur release calculated by the same means was considerably less, the authors suggest that their data indicate a minimum output and hypothesize that the eruption was capable of a substantial but short-lived climatic impact.

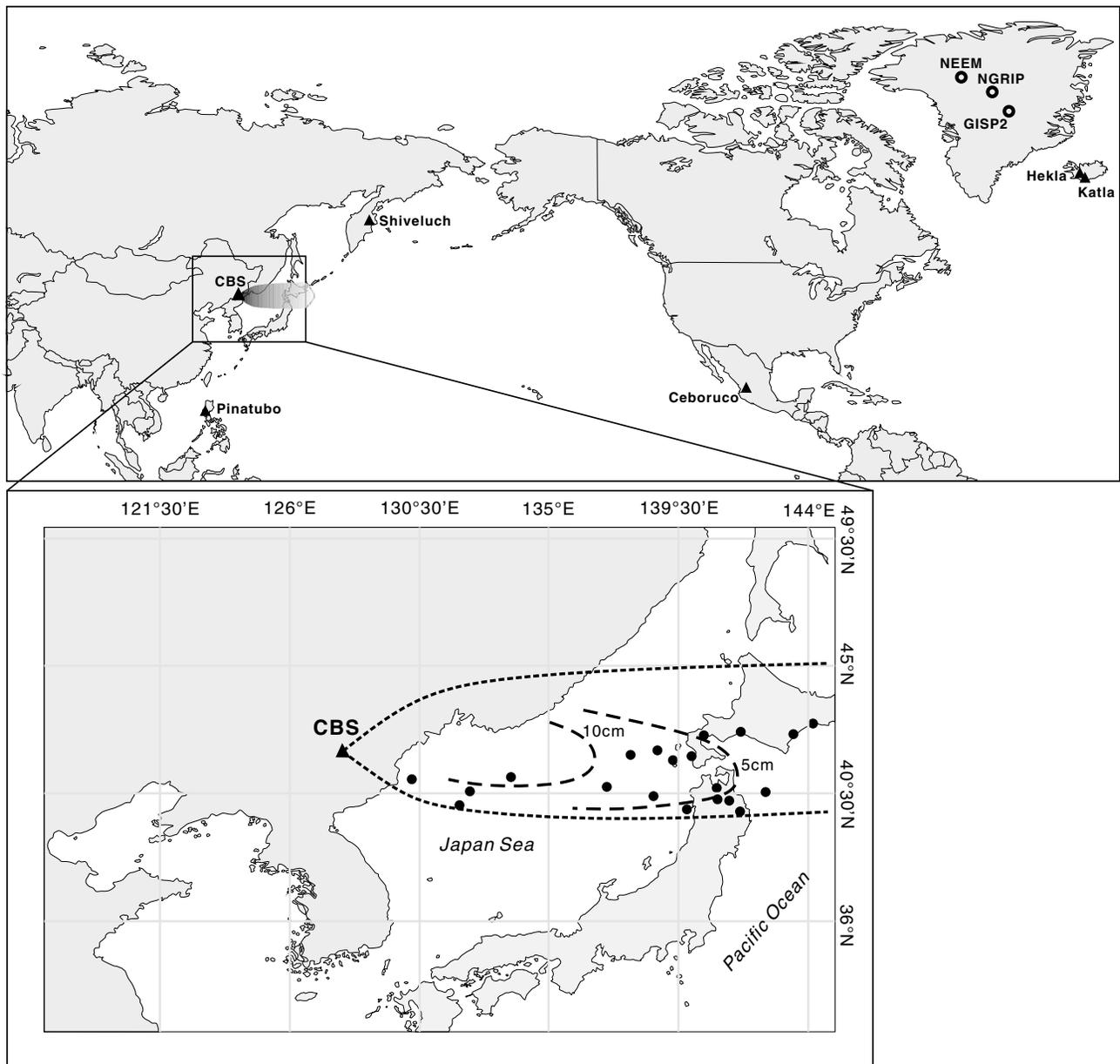


Figure 1. Overview map showing the location of Changbaishan volcano (CBS) and sites where Millennium eruption tephra has been reported. Distribution and thickness of distal tephra of this eruption are from *Machida and Arai* [1983]. Other occurrences of this tephra across the Japan Sea and Japan Island are from *Machida and Arai* [1983] and *Hughes et al.* [2013]. Locations of Eldgjá (Katla), NGRIP, NEEM-2011-S1, and other sites mentioned in the text are marked.

No unambiguous historical records of this eruption have been found in China or North Korea [*Machida et al.*, 1990]. The timing of the event was placed variably between the eighth and eleventh centuries according to conventional and wiggle match ^{14}C dating [e.g., *Horn and Schmincke*, 2000; *Nakamura et al.*, 2007]. More recent studies have attempted to refine the dating of the eruption through ^{14}C wiggle matching of proximal timbers buried by pumice fallout or pyroclastic flow from the eruption and have proposed dates of A.D. 923–939 [*Yin et al.*, 2012] and A.D. 940–952 [*Xu et al.*, 2013]. Through consideration of contemporary records, and tree ring and Greenland ice core data, these studies have reached divergent conclusions regarding its impact.

Historical records and tree ring indices from northeast Asia and Europe are consistent with dust veil events and associated cooling at ~A.D. 933/934 and ~A.D. 939/940 in Europe and Asia, each of which has been regarded as possible evidence for a protracted climate impact triggered by the Eldgjá eruption in Iceland

[Stothers, 1998; Fei *et al.*, 2003]. Correlation of the weather anomalies with the Eldgjá event has, however, been complicated by subtle discrepancies in proposed ice core dates for the event. In the Greenland Ice Sheet Project 2 (GISP2) core, sulfate and chlorine spikes at A.D. 938 ± 4 (GISP2 chronology) have been firmly linked with Eldgjá through the identification of tephra shards in the ice [Zielinski *et al.*, 1995]. Comparable aerosol spikes also attributed to the Eldgjá eruption in the Dye-3, GRIP, and North Greenland Ice Core Project (NGRIP) cores, on the other hand, are dated to A.D. 933 ± 1 (Greenland Ice Core Chronology 2005, GICC05) [Vinther *et al.*, 2006].

Differences in the ice core chronologies relate to the perceived annual layer counting errors within the cores; both chronologies are precisely dated in the midfourteenth century A.D. through the presence of tephra from Öraefajökull A.D. 1362 eruption [Palais *et al.*, 1991; Coulter *et al.*, 2012] and are further cross-matched using distinctive acid signals attributed to the historically dated Hekla A.D. 1104 and Vesuvius A.D. 79 events [Zielinski, 1996; Vinther *et al.*, 2006]. Barbante *et al.* [2013] have recently reported the identification of Vesuvius tephra in the GRIP ice core 1 year prior to the acid signal. Between these marker horizons, however, cumulative layer counting errors may exist and chronological precision is not absolute. Furthermore, sampling resolution in the GISP2 core is biennial, and this is factored into the dating error [Zielinski *et al.*, 1994].

For most researchers, subdecadal chronological imprecision is almost negligible, but for the correlation of ice core records to annually resolved or historical records, it can be critical, and the ice core volcanic record of the midtenth century A.D. is a case in point. Accepting the GICC05 age (A.D. 933 ± 1) for the Eldgjá eruption, Yin *et al.* [2012] argue for a second climatically effective event in that decade and suggest that documented weather and tree ring anomalies at A.D. 939/940 correlate with their proposed date of A.D. 938/9 for the Millennium eruption. Conversely, Xu *et al.* [2013] favor the GISP2 midpoint date of \sim A.D. 938 for the Eldgjá eruption and point to the absence of a sulfate spike and concomitant cooling in the GISP2 record in the period A.D. 940–952 to which they date the Millennium eruption; on this basis, they suggest that the Millennium eruption had a low sulfur injection into the stratosphere and that it thus had a negligible climate impact.

Recent research on the NGRIP ice core reported a bimodal population of unprovenanced tephra (QUB-1437) dated by GICC05 to \sim A.D. 940–941 \pm 1 [Coulter *et al.*, 2012]. Here we demonstrate that this tephra correlates on major element geochemistry with glass from the Millennium eruption, and we report the finding of the same tephra in the new NEEM-2011-S1 ice core from northern Greenland. Our results enable the possible atmospheric loading, and by extrapolation the climatic effectiveness, of the event to be evaluated critically.

2. Methods

The extraction and electron probe single-shard analysis of the NGRIP tephra QUB-1437 was described by Coulter *et al.* [2012]. On the basis of the sulfate record, three consecutive samples of NEEM-2011-S1 ice were cut from each of the intervals spanning \sim A.D. 939–942 and \sim A.D. 932–935 (GICC05 timescale) at the Desert Research Institute (DRI), Reno, and transferred to individual Nalgene bottles (supporting information). The meltwater was evaporated on frosted microprobe slides in a laminar flow cupboard at Queen's University Belfast (QUB), and the dry residues were covered with Buehler EpoxiCure resin. Tephra shards were quantified at 100X–400X magnification using a polarizing light microscope. Samples of white-yellow, grey, and black fallout pumice were collected from vertical exposures on the Korean flank of the Tianchi crater, reflecting progressive changes in the geochemical composition of the fallout; subsamples were mounted onto microprobe slides and covered with Buehler epoxy resin [Zhao, 2010]. Slides bearing tephra were ground with 12 μ m alumina and polished with 6 μ m, 3 μ m, and 1 μ m diamond paste to expose the surfaces of shards (distal material) or glass matrix (proximal material). The slides were carbon coated and glass composition was analyzed using the JEOL FEG-SEM microprobe at QUB under the same operating conditions as QUB-1437 [Coulter *et al.*, 2012]. A single analysis from one sample was obtained using the Cameca SX-100 at the University of Edinburgh (see supporting information for operating conditions). Secondary glass standards were measured at each analytical session to ensure instrument precision and accuracy (supporting information). Data have been normalized to 100% on a volatile- and water-free basis for plotting and statistical comparisons. Similarity coefficients for the tephras and possible correlates were calculated on the basis of major element analyses that comprised $>1\%$ concentration of the tephras, following Borchardt *et al.* [1972].

Procedures for the measurement of sulfate in NGRIP are described in Coulter *et al.* [2012] and the same methods were used to measure chloride. For NEEM-2011-S1, total sulfur, chlorine, and sodium concentrations

Table 1. Summary of Chemical Composition (Normalized to 100%) of Tephra Reported in This Paper (Proximal Material K From Changbaishan, QUB-1819 and QUB-1823 in NEEM-2011-S1) Along With Published Glass Data From Millennium Eruption Tephra From Northeast China and North Korea, Japan Sea and Japan, and Contemporary Tephra From the NGRIP (QUB-1437) and GISP2 (Glass B) Greenland Ice Cores

	Sample	SiO ₂	TiO ₂	Al ₂ O ₃	FeO	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	Cl	Analytical Total	
Changbaishan volcano	K-a (n = 6)	67.94	0.36	14.61	4.58	-	0.13	1.09	5.51	5.78	-	-	95.99	Mean
		1.31	0.15	0.47	0.39	-	0.14	0.36	0.14	0.15	-	-	0.50	2 SD
	K-b (n = 10)	74.88	0.20	10.66	4.05	0.08	0.02	0.26	5.32	4.52	-	-	96.34	Mean
		0.53	0.07	0.36	0.19	0.10	0.03	0.07	0.68	0.19	-	-	0.72	2 SD
	TC-a (n = 3) ^a	67.00	0.46	15.16	4.89	0.10	0.13	1.11	5.27	5.87	-	-	-	Mean
		1.33	0.12	0.55	0.25	0.11	0.08	0.17	0.46	0.32	-	-	-	2 SD
TC-b (n = 15) ^a	74.58	0.25	10.90	4.20	0.07	0.06	0.28	5.01	4.66	-	-	-	Mean	
	3.75	0.10	2.19	0.44	0.06	0.10	0.35	0.61	0.78	-	-	-	2 SD	
Japan region	B-Tm-a (n = 18) ^a	66.63	0.46	14.86	5.01	0.14	0.24	1.36	5.67	5.42	-	-	-	Mean
		2.48	0.39	1.00	0.67	0.20	0.42	0.88	0.64	0.41	-	-	-	2 SD
	B-Tm-b (n = 24) ^a	74.79	0.24	10.53	4.19	0.10	0.03	0.28	5.07	4.37	-	-	-	Mean
		1.94	0.17	1.04	0.32	0.10	0.07	0.16	0.54	0.47	-	-	-	2 SD
NGRIP	QUB-1437a (n = 10) ^b	68.05	0.30	14.86	4.80	0.14	0.12	1.26	4.69	5.76	-	-	95.77	Mean
		2.14	0.13	0.89	0.30	0.16	0.11	0.32	0.58	0.47	-	-	1.56	2 SD
	QUB-1437b (n = 6) ^b	75.58	0.20	10.41	4.18	0.08	0.04	0.29	4.57	4.65	-	-	96.42	Mean
2.87		0.12	0.76	0.23	0.09	0.07	0.35	1.22	0.43	-	-	1.17	2 SD	
NEEM-2011-S1	QUB-1819a (n = 6)	67.63	0.39	14.78	4.77	0.17	0.11	1.12	5.36	5.48	-	0.18	96.58	Mean
		1.50	0.02	0.97	0.42	0.19	0.08	0.36	0.86	0.36	-	0.08	1.86	2 SD
	QUB-1819b (n = 4)	74.74	0.22	10.57	4.07	0.04	0.02	0.29	5.04	4.54	0.00	0.46	96.18	Mean
		0.90	0.06	1.02	0.44	0.04	0.02	0.19	1.01	0.52	-	0.05	4.82	2 SD
	QUB-1819c (n = 1)	76.53	0.06	13.34	0.57	0.08	0.03	0.81	3.79	4.70	-	0.08	94.91	
QUB-1823a (n = 1)	71.29	0.46	14.91	2.17	0.08	0.40	1.57	2.71	3.72	-	0.13	92.49		
GISP2	QUB-1823b (n = 1)	77.94	0.02	13.49	0.51	0.19	0.05	0.78	4.04	4.74	-	0.11	94.23	
		69.2	1.3	13.5	5.3	0	1.1	3.2	2.9	3.6	-	-	-	Mean
	Glass B (n = 9) ^c	1.8	0.2	1.1	0.8	-	0.4	0.6	1.1	0.5	-	-	-	SD

^aMachida et al. [1990]; Zou et al. [2010]; Hughes et al. [2013].

^bCoulter et al. [2012].

^cZielinski [1996].

were measured simultaneously using a continuous flow analysis system coupled to two inductively coupled plasma–mass spectrometers with an effective sampling resolution of ~1 cm [McConnell et al., 2002]. Analyses were performed using 1 m longitudinal samples (30 × 30 mm) at the Ultra Trace Chemistry Laboratory at DRI. Non-sea-salt sulfur (nssS) concentrations were calculated from total sulfur concentrations using coregistered sodium concentrations as a sea-salt tracer and assuming a sulfur to sodium ratio in bulk seawater of 0.084 (i.e., [nssS] = [S] – 0.084[Na]). Here we report data from a secondary independent reanalysis of NEEM-2011-S1 (performed between A.D. 900 and 1130) for which the analytical setup was set to achieve increased depth resolution. Volcanic sulfate flux extraction follows procedures described by Sigl et al. [2013] with extracted deposition fluxes for volcanic events reproducing those from the primary analysis. The timescale was derived by annual layer counting constrained by volcanic signals [Sigl et al., 2013] identified in the NGRIP ice core record on the GICC05 timescale [Vinther et al., 2006].

3. Results

Tephra shards were identified in NEEM-2011-S1 samples dating to A.D. 940.5–941.4 (QUB-1819) and A.D. 933.8–934.5 (QUB-1823) (GICC05 chronology). QUB-1819 is characterized by mainly platy, colorless shards, the majority of which are less than ~10 μm in size; at least 30 shards between 10 and 50 μm in size were recorded (see supporting information for light microscope photographs). Analyses with acceptable analytical totals (>94%) were obtained from 10 shards. QUB-1823 contained only two shards, one a blocky, colorless shard (13 × 35 μm) and the other, small (10 × 6 μm) and slightly vesicular. Analytical totals were low (92–94%) for each shard but the results are sufficiently distinct to warrant reporting.

The results from QUB-1819 and QUB-1823 are summarized in Table 1 along with newly analyzed proximal material from Tianchi (K), QUB-1437 from NGRIP, published proximal and distal data from Millennium eruption deposits in NE China and Japan (TC, B-Tm), and the dacitic component (Glass B) found with Eldgjá basaltic tephra in GISP2 (see supporting information for full data sets and similarity coefficients). The proximal and distal Millennium eruption glass data indicate a bimodal (rhyolitic and trachytic) magmatic composition that is clearly distinct from other contemporary large Northern Hemisphere eruptions (Figure 2); of particular

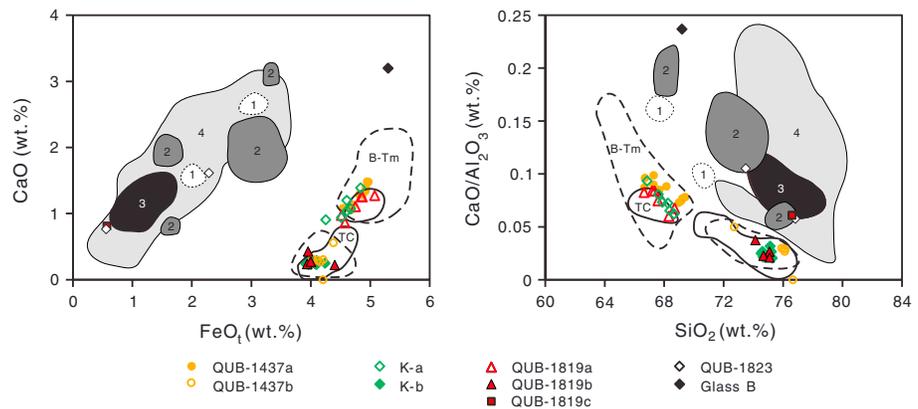


Figure 2. Binary plots demonstrating major element chemical similarity of glass shards from Greenland ice cores with both proximal (Changbaishan volcano) and distal (Japan areas) deposits of the Millennium eruption tephra. Glass compositions of K, QUB-1437, QUB-1819a, and QUB-1819b are comparable to those of published proximal and distal data produced by the Millennium eruption and clearly distinct from broadly contemporary eruptions with similar compositions, such as (1) the Jala pumice of Volcán Ceboruco, Mexico [Gardner and Tait, 2000], (2) Hekla A.D. 1104 (data from <http://www.tephrabase.org/>; sources Boyle [1994]; Larsen et al. [1999]), (3) Holocene tephra of Shiveluch volcano, Russia [Ponomareva et al., 2007], and (4) widespread Quaternary tephra around Japan [Aoki and Machida, 2006] and the dacitic Glass B in GISP2 at A.D. 938 ± 4 [Zielinski et al., 1995] (envelopes represent distribution of published data points). Published B-Tm and TC data are from Machida et al. [1990]; Zou et al. [2010], and Hughes et al. [2013], and QUB-1437 data are from Coulter et al. [2012]. QUB-1823 is clearly distinct from GISP2 Glass B.

note is that the ratio of FeO_t to CaO in the Millennium eruption magma is greater than in products from other volcanic regions. QUB-1437 and QUB-1819 reveal the same distinctive bimodal character, and provide compelling evidence for the arrival of Millennium eruption ash in Greenland. QUB-1819 also contains an outlying analysis (QUB-1819c) characterized by a low iron and lesser alkali content that is not consistent with volcanic products from Iceland, Kamchatka, or China (G. Larsen, personal communication; V. Ponomareva, personal communication, 2013). The two shards from QUB-1823 are also bimodal, and do not correlate with Eldgjá, GISP2 Glass B, or any other Icelandic eruption at this time. Their provenance is not currently known. The rhyolitic shard (QUB-1823b) is, however, comparable in major element composition to QUB-1819c.

4. Discussion

Our tephra results from NEEM-2011-S1 do not enable us to confirm that the A.D. 933–934 (GICC05) acid spike was the result of the Eldgjá eruption, but given the petrological data from Eldgjá deposits [Thordarson et al., 2001] and the similarities in the chemical records between GISP2 and other Greenland ice cores, it is reasonable to argue that this event at least partly (and perhaps mainly) contributed to the volcanic signal in Greenland ice at ~A.D. 933/934 (GICC05) or ~A.D. 938/939 (GISP2 chronology). In the absence of a tephrochronological confirmation of a common source of these acid layers, the exceptional high chlorine concentrations accompanying the volcanic signal provides an additional stratigraphic link between the three ice core records, as well as with the Dye-3 and GRIP ice cores [Clausen et al., 1997]. Nevertheless, the analyses from QUB-1823 raise the possibility that one or more other eruptions also occurred at this time and, notwithstanding uncertainty regarding the event’s precise date, may have augmented the climate impact inferred from historical records [Stothers, 1998; Fei et al., 2003].

The unambiguous presence of Millennium eruption tephra in NGRIP and NEEM-2011-S1 enables a precise correlation of the event with synchronous, short-lived (<1 year) sulfate spikes at A.D. 941 ± 1 (GICC05), and clearly places the eruption ~7 years after the acid peak (sulfate and chlorine) attributed to Eldgjá (Figure 3). In addition, the strong seasonality expressed in the natural background of the plotted chlorine/chloride data provides a means to visualize the data underlying the annual layer counting in NGRIP and NEEM-2011-S1 and supports our attributed age offset of ~7 years between the Eldgjá and Millennium eruptions. The apparent absence of an acid signal in GISP2 in the A.D. 940 s may be due to the lower resolution (~2 years) of this core’s data over this interval [Zielinski et al., 1994]. The Millennium eruption is thus dated to either ~A.D. 941 ± 1 (GICC05) or ~A.D. 945 ± 4 (GISP2 chronology), and within the dating error of the ice core chronologies is unlikely to have occurred any earlier than the winter of A.D. 939/940. Ostensibly, historical observations

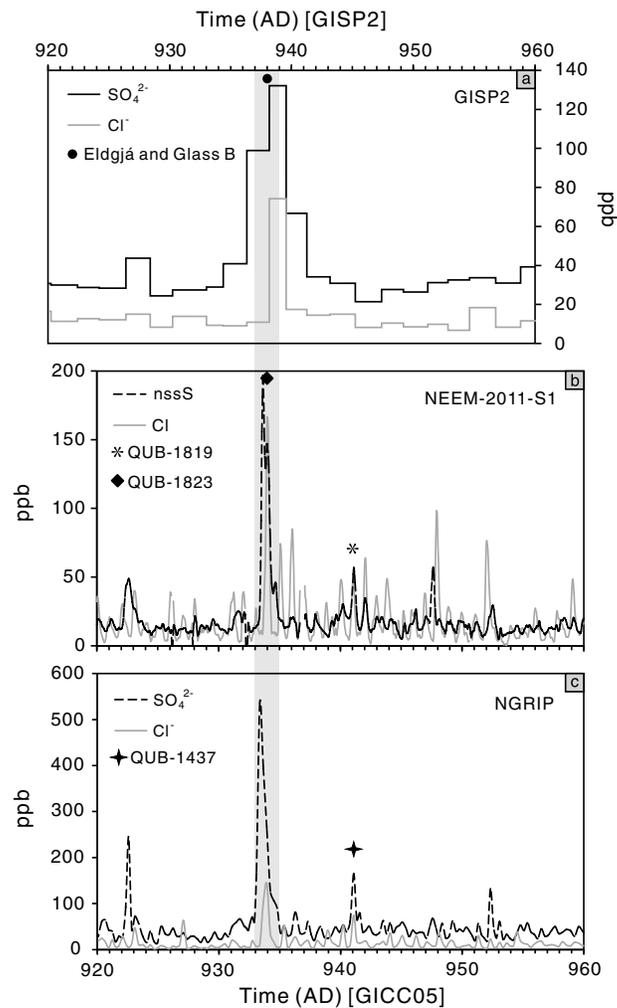


Figure 3. Cross comparison of chemical data from NGRIP, NEEM-2011-S1, and GISP2 ice cores for the period A.D. 920–960: (a) total sulfate and chloride concentrations in GISP2, (b) non-sea-salt sulfur and chlorine concentrations in NEEM-2011-S1, and (c) total sulfate and chloride concentrations in NGRIP. Plots are aligned on the basis of the common volcanic signal attributed to the Eldgjá eruption (grey band). GISP2 data are from <http://www.ncdc.noaa.gov/paleo/icecore/greenland/summit/document/gispdata.htm>. NGRIP sulfate data are from Coulter *et al.* [2012], converted to ppb scale. Sampling resolution of GISP2 is biennial and NGRIP is four samples a year (5 cm resolution). NEEM-2011-S1 resolution is 20 samples per year (1 cm resolution). NGRIP and NEEM data from the sections shown are provided in supporting information.

consistent with a large regional eruption—unusual sonic effects (“thunders like a drum”) in Japan and Korea and “white ash rain” in Japan in the winter of A.D. 946–7 [Xu *et al.*, 2013]—favor the later, GISP2 age estimate. Uncertainty regarding the precision of the ice core chronologies prohibits a definitive linkage or distinction of the event from historically recorded and tree ring dated events. Nevertheless, the potential climate impact of the eruption can be assessed using aerosol data from the ice cores.

QUB-1437 and QUB-1819 are each directly associated with distinct peaks in volcanic sulfates (Figure 3). In NEEM-2011-S1, QUB-1819 and a sulfur spike occur amidst an enhanced volcanic signal beginning 2 years prior to the Millennium tephra level, suggesting that more than one eruption is recorded in the ice at this time, a prospect supported by the outlying shard QUB-1819c. The coeval deposition of Millennium eruption tephra and aerosols over Greenland implies rapid initial transport to the ice sheet through a common tropospheric or stratospheric pathway. The dispersal pattern of the Millennium eruption tephra [Machida and Arai, 1983] also indicates predominant east-northeastward upper winds during this eruption (Figure 1) that would have favored rapid transport of ash and aerosols toward Greenland within weeks or months of the eruption. Sulfate deposition estimates at this time are in the order of 11 kg km^{-2} in NGRIP and 13 kg km^{-2} in NEEM-2011-S1 [Sigl *et al.*, 2013].

The higher resolved sulfate record from NEEM-2011-S1 deployed for this study further suggest that 9 kg km^{-2} were deposited during the peak associated with tephra (247.29–247.13 m) while an additional 4 kg km^{-2} were deposited during the earlier event. These levels are considerably less than the estimated deposition following eruptions such as Tambora 1815 ($\sim 40 \text{ kg km}^{-2}$) or Krakatau 1883 ($\sim 15\text{--}18 \text{ kg km}^{-2}$) but are consistent with the modest, petrologically estimated sulfur output calculated by *Horn and Schmincke* [2000] for the Millennium eruption. We conclude, therefore, that the stratospheric loading from this event is unlikely to have been sufficient to impact significantly on extraregional climate. Furthermore, growth-ring data from trees killed by the fallout deposits indicate that the eruption occurred sometime between autumn and spring [Yin *et al.*, 2012], when the climate impact of eruptions in higher latitudes may be negligible [Kravitz and Robock, 2011].

5. Conclusions

Tephra and chemical analyses from the NGRIP and NEEM-2011-S1 Greenland ice cores have enabled an assessment of the likely atmospheric aerosol loading of the Millennium eruption and an evaluation of the extraregional climate impact of the event. Although the eruption was sufficiently powerful to disperse tephra to northern Greenland, associated non-sea-salt sulfur and sulfate concentrations are low and short-lived. This finding suggests that the eruption would not have had any substantial climate impact. The exact timing of the eruption remains ambiguous because the ice core chronologies are not absolutely dated at this time but in light of the relatively low sulfate loading from the event as well as an expected time lag in European climate response to volcanic forcing [Esper *et al.*, 2013], weather anomalies across Eurasia in A.D. 939–940 (mainly concentrated in Europe) are unlikely to be the result of this eruption. On the basis of a stratigraphic (~ 7 years) offset, we can nevertheless distinguish the Millennium eruption from the major Eldgjá eruption which remains a contender for a volcanic source of the climate perturbations at A.D. 939–940.

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