

**Smoking guns and volcanic ash: the significance of sparse tephras in Greenland ice cores**

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## Abstract

Volcanic ash (fine-grained tephra) within Greenland ice cores can greatly complement the understanding of past volcanism, and its environmental and societal impacts. The presence of ash in sparse concentrations in the ice raises questions about whether such material represents primary ashfall in Greenland or resuspended (remobilised) material from continental areas. Here, we investigate this issue by examining tephra content in quasi-annual samples spanning a ~20-year period from two Greenland ice cores, and considering their relationships to sulphur and particulate data from the same cores. We focus on the interval 815–835 CE as it encompasses a phase (818–822 CE) of heightened volcanogenic sulphur previously ascribed to an eruption of Katla, Iceland. We find that tephra is a frequent but not continuous feature within the ice, unlike similarly-sized particulate matter. A solitary ash shard whose major element geochemistry is consistent with Katla corroborates the attribution of the 822±1 CE sulphur peak to this source, clearly showing that a single shard can signify primary ashfall. Other tephras are present in similarly low abundances but their geochemistries are less certainly attributable to specific sources. While these tephra shards tend to coincide with elevated sulphur and fine (<10 µm) particulates, they are not associated with increased coarse (>10 µm) particle concentrations that might be expected if the shards had been transported by dust storms. We conclude that the sparse shards derive from primary ashfall and we argue that low shard abundance provides insufficient grounds to dismiss the potential significance of sparse tephras.

**Keywords:** Greenland ice cores; Katla; Sparse tephras; Resuspended volcanic ash; Dust storms; Volcanic eruptions

**Running head:** Sparse tephras in Greenland ice

## Introduction

In recent years, evidence for past volcanic impacts on climate variation and societal transformation has increasingly been posited (Esper et al. 2013; Sigl et al. 2015; Stoffel et al. 2015; Büntgen et al. 2016). The climate effectiveness of eruptions stems from the capacity of material injected into the stratosphere, particularly sulphur-containing gases that convert to sulphate aerosols, to deflect incoming solar radiation over the course of weeks to years, thus triggering surface cooling (Robock 2000), although ash particles may offset the cooling in some instances (Flanner et al. 2014). Volcanic eruptions vary greatly in their magnitude, duration and type of emission, and their latitudinal location is also a significant factor in the potential extent of their atmospheric impact (Toohey et al. 2019). As volcanic eruptions occur globally on a continual basis, including several each decade that penetrate stratospheric levels, it remains to be established which types of events pose the greatest risk to the climate system.

The understanding of past volcanic impacts hinges substantially on the polar ice core records that present through their chemistry an extended history of volcanic activity, most notably of eruptions that emitted sufficient aerosols to have potentially impacted the Earth's radiative balance (Hammer et al. 1980; Zielinski et al. 1994; Sigl et al. 2013). For the Late Holocene, ice cores offer a level of chronological precision that enables their records to be reconciled with historical events, thus allowing climate perturbations and any ensuing societal impacts to be evaluated (Sigl et al. 2015; Büntgen et al. 2016). Identifying the source eruption is key to deciphering the volcanic processes that impact the climate system. For example, atmospheric loading of volcanic aerosols is best reconstructed if the latitude, other eruption parameters (e.g., plume height, eruption style) and season are known. However, in many regions, observational records of eruptions began only in recent centuries and volcanic histories remain very poorly resolved in many regions (Siebert et al. 2010). Furthermore, secure attribution of volcanic signals to specific eruptions can only be achieved through the analysis of associated volcanic ash (fine-grained tephra) particles whose glass geochemical composition may point to the source eruption, volcano or volcanic region.

Transport of volcanic ash to polar regions is constrained by several factors, however, not least injection height, meteorological conditions at the time of the eruption over both the source volcano and the deposition site, and general atmospheric circulation patterns. Given these limitations, it is perhaps not surprising that concentrations of ash particles large enough (usually  $>20\ \mu\text{m}$ ) to be geochemically-typed using standard tephra-analytical techniques in polar ice are frequently low (Iverson et al. 2017). For Late Holocene Greenland ice, Coulter et al. (2012) examined multi-annual samples with as few as one tephra shard identifiable using light microscopy, while other studies have been limited to analysing particles  $<10\ \mu\text{m}$  using sub-optimal techniques for geochemical characterisation (Palais et al. 1992; Zielinski et al. 1997; Barbante et al. 2013). Such very fine-grained material might not be recognisable as volcanic glass using standard optical microscopy for routine tephra screening.

A major concern relating to sparse cryptotephra in polar ice is whether they represent primary ash-fall or reworked (secondary) material. Prevailing wind patterns, the high latitude and altitude of the ice core sampling locations, and the specific environmental conditions required to resuspend ash from older deposits are amongst the leading factors that mitigate the incorporation of reworked tephra into the ice. Nevertheless, it is conceivable that strong dust storms could transport resuspended sediment, including tephra, over long distances to polar areas. Aeolian resuspension of ash is especially likely in areas where pedogenesis and stabilisation by vegetation cover is retarded or absent, namely in very cold or very arid regions. In the Valley of the Ten Thousand Smokes, Alaska, for instance, tephra beds from the 1912 Novarupta eruption remain exposed as climate and topography combine to hinder the establishment of pioneering vegetation (Hildreth & Fierstein 2012). Under certain wind conditions, ash from these beds can be resuspended and elevated as much as 3 km into the atmosphere, equivalent to a plume from a Vulcanian-style eruption and sufficient to enable ash to be dispersed over several thousand kilometres (Hadley et al. 2004). How then do we differentiate primary ashfall from dust deposition events incorporating remobilised tephra? While reworked glass can sometimes be evidenced by rounded edges, corrosion or altered

major element geochemistries (Lowe 2011), these indices may depend upon factors such as the length of time a tephra has been exposed to weathering prior to remobilisation, or transport mechanisms. Low analytical totals in geochemical analyses can also result from post-deposition hydration of the glass under exposed conditions, although other factors contributing to low totals include thin glass walls and instrumental error (Froggatt 1992; Hunt & Hill 1993).

In terms of the potential source area of reworked volcanic material reaching Greenland, we might reasonably assume a correlation with the sources of fine particulate matter in the ice that regularly reaches polar locations. Strontium (Sr) and Neodymium (Nd) isotope analyses of  $<5\ \mu\text{m}$  particles in high-altitude Greenland ice cores indicate a predominantly east Asian origin (Bory et al. 2002; Bory et al. 2003), although others propose that a north African source may be underestimated in these studies (Tanaka & Chiba 2006; Lupker et al. 2010). Winter atmospheric circulation over both the Greenland NEEM and Tunu ice-core drilling sites are dominated by the polar vortex. In spring to summer, air masses over both sites originate mainly from the west but both sites are also influenced by air masses from the North Atlantic, arriving at NEEM from the south and at TUNU from the northeast and southeast (Steen-Larsen et al. 2011; Maselli et al. 2017). With the exception of Iceland and the Jan Mayen islands (whose tephra are geochemically well-characterised), however, these regions do not feature unconsolidated ash beds from which reworked tephra could derive. In Iceland, dust remobilisation also occurs predominantly in spring and early summer and, from southern Iceland at least, dust is mainly transported southwards (Prospero et al. 2012). On balance, it seems that the most probable sources of reworked tephra that might reach Greenland will lie to the west.

How meaningful, therefore, are sparse tephra particles recorded in the ice? Do occasional shards merely reflect a background signal of remobilised ash particles or can they represent primary ashfall? To address these questions, we consider a ~20-year interval of ice from two northern Greenland ice cores – NEEM-2011-S1 and TUNU2013 (Fig. 1) – to investigate the frequency and origin of volcanic ash in the records, and their relationship to other volcanogenic proxies.

## Materials and methods

Sulphur concentrations from the TUNU2013 and NEEM-2010-S1 Greenland ice cores have previously been reported (Sigl et al. 2013; Sigl et al. 2015). For the TUNU2013 core, fine particulate matter was measured for two fractions (2.6–4.5  $\mu\text{m}$ , 4.5–9.5  $\mu\text{m}$ ) during continuous flow analysis of an adjacent parallel melter stick from the core (Ruth et al. 2003; Sigl et al. 2013; Jensen et al. 2014). We examined sections of ice from both cores spanning the period ~815 to ~835 CE, encompassing an extended volcanogenic acid signal between 818 and 822 CE (Büntgen et al. 2017). Constrained at the 774 CE cosmogenic event (Miyake et al. 2012), both chronologies have an error of  $\pm 1$  year in this interval (Sigl et al. 2015). In total, 4.42 m of ice from NEEM-2010-S1 and 2.01 m from TUNU2013 were subsampled at approximately annual resolution, comprising 40 individual samples. Samples were cut from archived ice at the Desert Research Institute, Reno, using a bandsaw and transferred to sterile Nalgene bottles. These samples comprise surface areas mainly between 15–20  $\text{cm}^2$ , greater than the area sampled in previous studies (e.g. Coulter et al. 2012); coupled with the high-latitude, high-elevation locations of the coring sites, they present only a small opportunity to capture remnants of sparse or fleeting ash clouds. In addition, a sulphur peak at 822 CE in TUNU2013 was subsequently resampled with respect to a prominent peak in particulate matter; for this event, fine resolution (sub-annual) samples were collected immediately before, across and after the particle peak, bringing the total number of samples examined to 43.

Samples were shipped to Queen's University Belfast where they were prepared by centrifuging the melt-water to concentrate the particulates, which were then pipetted onto pre-ground glass slides in a laminar flow cupboard and covered in Buehler ExopyCure2 resin. Each sample was assigned a unique identifier code (prefixed QUB-). Slides were scanned at x100 magnification on a light microscope and tephra particles were identified with the aid of cross-polarised light. The number of particles  $>10 \mu\text{m}$  within each sample was also counted and converted to concentrations to quantify

coarse material flux within the ice cores. For samples containing tephra, thin-sections were prepared by grinding and polishing the slides until the surfaces of tephra shards were freshly exposed. Major element geochemical composition of the glass shards was determined using integrated electron and wavelength dispersive spectrometry on a JEOL FEGSEM 6500F at Queen's University Belfast following Coulter et al. (2012). Secondary glass standards were analysed at the start and end of each session and between samples to evaluate analytical precision. All data were normalised to facilitate comparison with published data, and are reported, along with secondary glass standard data, in Supplementary Dataset 1.

## Results

Fig. 2 illustrates the positions of the tephra samples relative to the available sulphur and particle concentrations in the NEEM-2011-S1 and TUNU2013 ice cores. The extended period of heightened sulphur volcanism evident in the NEEM-2011-S1 core between 818–822 (Büntgen et al. 2017) is also visible in TUNU2013. The TUNU2013 record contains greater inter-annual variability, however, potentially owing to 1) the coring site's closer proximity to Iceland, 2) the site's lower elevation that places it closer to tropospheric sulphur emission sources, and 3) the lower snow accumulation rate which allows for enhanced surface snow mixing. Fluctuations in the TUNU2013 fine ( $<10\ \mu\text{m}$ ) particle fractions follow neither an annual nor seasonal pattern. Both fractions show close correspondence with each other, with elevated levels at 814, 817, 822, 823, 825 and 830 CE, with smaller peaks at 815, 816, 819, 821, 826, 831 and 832 CE. Visual examination of the samples under a light microscope showed the prominent peak at ~830 CE (QUB-1886) to relate to mineral dust. The smaller peaks at ~826 CE (QUB-1883), 831 (QUB-1888) and 832 CE (QUB-1889) also contained mineral dust, and variously included occasional examples of charcoal, fungal spores and diatoms. Coarse particles ( $>10\ \mu\text{m}$ ) were present in all samples, but their concentrations were an order of magnitude greater in the TUNU2013 core (Supplementary Dataset). Notable increases in coarse particles occur in the NEEM-

2011-S1 core at 828–9 (QUB-1847) and 835–6 CE (QUB-1853), and in TUNU2013 at 814 (QUB-1870) and 826–7 CE (QUB-1883); only the last-mentioned peak coincides with elevated sulphur levels. QUB-1878, dating to 821–2 CE, contained a large number of poorly preserved sponge spicules and diatoms, as did the overlapping sample QUB-1974; the presence of such material implies the transport of marine water to the sampling site.

Only five of the 43 quasi-annual samples yielded unambiguous tephra particles, ranging from one to five shards per sample (Supplementary Dataset; Supplementary Information Figures 1–6); in some of these, additional potential shards were present but were too small to enable certain identifications. A probable glass shard was recorded in one other sample (QUB-1841; Supplementary Information Fig. 2) but its diminutive size prohibited geochemical analysis. Although it is now possible on certain microprobes to analyse tephra using a 3  $\mu\text{m}$  focused beam (Hayward 2012), it remains a major challenge to expose a large enough surface area of glass in thin section without destabilising the resin or polishing away the shard, especially if shards have a platey (i.e., thin) morphology. Possible tephra shards were also present in QUB-1870 but none of the analysed targets proved to be glass; this sample also contained the highest concentration of coarse particles in all of the ice examined in this study. As a test of the reliability of the analytical data obtained from the very sparse shards and to ensure that results were not skewed by the presence of mineral inclusions, multiple analyses from each shard were obtained where particle size permitted.

Analytical totals frequently fell below the normally accepted threshold of 95 wt% proposed by Hunt & Hill (1993), albeit above ~90% wt% deemed acceptable by Froggatt (1992). We have found this to be a frequent challenge when working with small particles as epoxy resin is entrapped during analysis, demonstrated by elevated Cl (Coulter et al. 2012). In some instances reported here, we attribute low totals at least in part to instrumental settings, noting lower than usual totals for the secondary glass standards at the time of analysis (see Supplementary Dataset).



*QUB-1835, 815 CE*

A single bubble-walled shard of colourless glass in NEEM-2011-S1 yielded three analyses but the composition of the initial analysis is not replicated by the subsequent two. Geochemical heterogeneity and low analytical totals (<95 wt%) highlight a need for caution in the interpretation of the data. However, the glass showed no physical signs of weathering and post-analysis examination reveals burning beyond the edges of the shard (Supplementary Information Fig. 1), which will have contributed to low totals, as well as Cl entrapment. All things considered, therefore, we attribute the low totals to problematic analyses rather than hydration. An attempt to characterise the trace element composition of the QUB-1835 shard using Laser Ablation Inductively Coupled Plasma Mass Spectrometry proved unsuccessful due to the return of a low signal, reflecting the limited glass available for analysis. In NEEM-2011-S1 at this time, sulphur remains at background levels.

Amongst our database, the major element composition of the first analysis most closely matches that of Lipari, Italy (Supplementary Information Fig. 7), the Monte Pilato eruptive event of which has been dated to 776 +100/-90 CE (Keller, cited in Lucchi et al. 2007) and is therefore within the same timeframe. The subsequent analyses are not, however, consistent with this source, but are instead similar to some material from the South Mono eruption of Mono Craters, USA, dated to 594-648 CE (Bursik et al. 2014). Interestingly, Bursik et al. (2014) have posited a remobilisation event following a fire ~200 years after the South Mono eruption. However, we observe no obvious increases in either coarse particle content or charcoal that might suggest a dust deposition event, particularly one associated with a fire. No tephra was identified in the corresponding levels in TUNU2013.

All in all, given the questionable quality of the data, it is not possible to propose correlatives for this tephra, and further sampling of ice of this age is warranted. The geochemical heterogeneity of the shard is possibly indicative of secondary deposition of weathered glass, but in view of the shard size (>40  $\mu\text{m}$ ), the absence of visible signs of weathering, and the lack of other material in the sample

that would point to a dust event (e.g., charcoal, pollen, coarse-grained minerals), we conclude that QUB-1835 is likely to be the result of primary airfall.

#### *QUB-1872, 816 CE*

Three analyses were obtained from a colourless, platey shard in TUNU2013. The shard is characterised by a very distinctive high-SiO<sub>2</sub> and very low FeO<sub>total</sub> and CaO content. Analytical totals were consistently low, but the data indicate a homogenous chemistry across the shard. We have been unable to identify a possible source for this tephra, but similar tephras of Pliocene to Pleistocene age have been reported from marine cores from the northwestern Pacific (Cao et al. 1995), suggesting a potential derivation from the Kamchatkan/Kurile region. Four additional tephra-like particles measuring ~10–20 µm were present but were too small to analyse on our instrument. A range of similarly-sized minerals, including feldspars, were present but not notably abundant. The TUNU2013 sulphur and particle records are slightly elevated in early 816 CE. The potential source region (i.e., upwind of Greenland) for the analysed shard increases the likelihood that this material could be reworked, yet the homogenous glass geochemistry and the simultaneous increase, albeit modest, in sulphur are consistent with primary deposition following an eruption.

#### *QUB-1873, 817 CE*

A single shard of brown, cusped glass produced three consistent analyses that indicate a trachydacitic composition. No certain correlatives were identified for this tephra, but the chemistry is similar to the ~80 ka-old NEEM2049.3 identified in Greenland ice and attributed to the Eastern Aleutian Arc (Bourne et al. 2016; Supplementary Information Fig. 8). It also bears some similarity to trachydacitic glass from Shiveluch (Ponomareva et al. 2015; Ponomareva et al. 2017) though such glass has not been reported from this time-interval. Some smaller glass particles (<20 µm) were also noted and other large particles were present in below-average concentrations. The sample's

position in TUNU2013 coincides with small peaks in sulphur and fine particles in early 817 CE. On the basis of the consistent analyses and association with volcanogenic sulphur, we propose that this particle is from primary ashfall.

#### *QUB-1974, 822 CE*

Only one analysis was feasible on the isolated light brown, cusped shard recorded in this TUNU2013 sample. Despite a low analytical total, the distinctive geochemistry can reliably be attributed to Katla, Iceland (Supplementary Information Fig. 9). This sample is directly associated with major fine particle and sulphur peak in early 822 CE, but coarse particles were not notably abundant in this or the overlapping sample QUB-1878. In view of the shard's unambiguous geochemistry and its direct association with a significant sulphur peak, we conclude that the shard is undoubtedly from primary ashfall.

#### *QUB-1880, 824 CE*

Of the four certain glass shards identified in this TUNU2013 sample, analyses were obtained from three. All three shards are rhyolitic and characterised by low CaO (<0.9 wt%) and high K<sub>2</sub>O (>4 wt%). One shard yielded three slightly dissimilar analyses, all comparatively enriched with Al<sub>2</sub>O<sub>3</sub>, FeO<sub>total</sub> and TiO<sub>2</sub> pointing to entrapment of a large mineral inclusion that is visible within the glass (Supplementary Information Fig. 6a). The remaining two shards show a strong correlation with material identified by Nooren et al. (2009) and Nooren et al. (2017) amongst El Chichón samples in delta deposits to the north of this volcano in Mexico. Although Nooren et al. (2009, p. 109) describe their Unit E shards as sharp-edged, the geochemical similarity of this material with 84 ky-old Los Chicoyos tephra from Atitlán, Guatemala, led Post (2016) and Nooren et al. (2017) to conclude that this component represents older, reworked tephra rather than primary ashfall. Similar cryptotephra has been reported in sparse quantities from European terrestrial sediments dating to approximately

650 CE and 400 CE (Plunkett & Pilcher 2018: DOM-3 and DOM-4 Group 1) and NEEM-2011-S1 ice dating to 536 CE (Sigl et al. 2015: QUB-1859 shards 1, 10 and 11). Contrary to Loveluck et al. (2018), this material is very distinct from Icelandic tephra and bears no geochemical similarity to the shards reported from the Colle Gnifetti ice core from Switzerland. The geochemical homogeneity of the distal correlatives leads us to question the attribution to a Pleistocene eruption of Atitlán, if indeed the cryptotephra are from the same source, as we would expect greater evidence for weathering from an 84 ka-old deposit. Furthermore, it seems unlikely that remobilised sediment from a tropical environment would be transported to northern Europe and Greenland. Considering the frequency of similar tephra in Mexican sediments, a source within this broader region seems likely.

The sample partly captures a minor particle peak in mid-823 CE but coarse particle concentrations are below average for this ice core section. Despite a lack of association with sulphur, the shards comprise sharp glass and, with the exception of the example containing a mineral inclusion, have yielded consistent analyses. We argue, therefore, that they derive from a primary ashfall of unknown origin, but possibly emanating in southern North/Central America.

## **Discussion**

Our study of two continuous ~20-year sections of Greenland ice reveals that tephra presence in Late Holocene ice is not constant. While the low flux of optically-identifiable volcanic particles reaching the ice cores may imply that isolating tephra in these records is rather haphazard, a correspondence between observed glass shards and elevated fine particle content demonstrates that these sparse occurrences are associated with pulses of non-seasonal dust reaching the ice. In contrast, coarse minerals in all of our samples demonstrate that particles in the size range of the tephra shards we report are regularly transported to Greenland, especially to the northeastern site of Tunu, but they are not palpably correlated with elevated sulphur or tephra (Supplementary Information Fig. 11). Hence, we can conclude that while large particles are persistently present in the ice cores, tephra

shards are not, nor are shards associated with elevated levels of coarse material. We infer, therefore, that the sparse tephras we record are unlikely to be indicative of dust storm events.

Of the five cryptotephra samples described here, we can identify with certainty the provenance of only one. Fortuitously, the unique geochemical composition of Katla basaltic tephra enables us to attribute QUB-1974 with high confidence to this volcano. Büntgen et al. (2017) demonstrated that a Katla eruption triggered a glacial outburst that is dated by dendrochronology to 822/3 CE, and proposed that this event was responsible for a sulphate anomaly in Greenland ice cores at this time. Our tephra shard, coinciding precisely with the 822 CE sulphate and particle peaks in TUNU2013, unequivocally corroborates this correlation, and we conclude that this solitary particle represents primary ashfall. The coeval deposition of sulphur and glass implies simultaneous transport by tropospheric or stratospheric winds, rather than the subsequent remobilisation of glacial flood deposits as described by Prospero et al. (2012).

QUB-1835 has produced heterogenous results from a single shard, preventing firm attribution to any specific source. The disparity may signify glass alteration, and there is no associated rise in volcanogenic sulphur to signify a coeval eruption. The absence of a sulphur peak in the ice does not, however, exclude the possibility that the material derives from primary ashfall: Öraefajökull 1362 CE tephra was recorded in high concentration in the GRIP core in ice of the same age but was not accompanied by a sulphate layer (Coulter et al. 2012). Perhaps more telling is the absence of similarly-sized dust particles and other terrestrial content that one might expect to be remobilised with reworked tephra. The remaining three samples in this study are more robust in their geochemical characterisation but nevertheless pose conundrums in terms of understanding their derivation. QUB-1872 and -1873 both possibly originate from the North Pacific region but neither clearly corresponds with a known Late Holocene tephra. In both instances, the potential source region lies upwind of Greenland and it is conceivable that the shards represent reworked material, the geochemistry of which might have been altered through weathering. However, the coincidence

of elevated sulphur and the absence of increases in other coarse particles strengthens the likelihood that the shards derive from unidentified eruptions.

QUB-1880 comprises a larger number of shards whose origins are less clear; that the major element geochemical composition was reproducible from two shards suggests that the glass geochemistry is unaltered. We find that material with matching major element geochemistry was deposited in Greenland and continental Europe on multiple occasions in the centuries preceding 824 CE, although the concentrations of tephra associated with these events is generally very low. We note a possible correlative also with an outlying population reported amongst El Chichón tephra in Mexico, implying a possible origin in southern North America or Central America, an improbable source region of remobilised dust that could reach northern Greenland and Europe. We propose, therefore, that the multiple incidents of this glass type represent primary ashfall.

Despite the difficulties in determining the source of all but one of the glass shards we have analysed, we conclude – with varying degrees of confidence – that the material represents primary deposition. We find no certain evidence of physical or chemical alteration of glass, and the samples containing confirmed tephra are not associated with increases in similarly sized terrestrial particles that we might expect had sediment been remobilised in dust storms.

## **Conclusion**

We have examined the occurrence of tephra in two Greenland ice cores spanning a period of ~20 years, and their relationship to volcanogenic sulphur, fine (2.5–9.5  $\mu\text{m}$ ) and coarse (> 10  $\mu\text{m}$ ) particulate matter. Like previous studies (Abbott & Davies 2012; Coulter et al. 2012), we find that not all sulphur peaks are associated with tephra: this may be due to eruptions that produced little or no ash, or ash clouds not reaching the ice core localities due to meteorological conditions or the distance of the eruption from the polar region. Equally, tephtras are not always associated with

346 sulphur peaks, highlighting that not all volcanic eruptions emit aerosols in concentrations to impact  
347 climate.

348 Tephra was present in six (possibly seven) of our 43 samples in low concentrations, despite the  
349 relatively sizeable samples examined in this study. These findings demonstrate that tephra  
350 occurrence in the ice cores is not rare, but nor is it consistently present as a background input as  
351 similarly coarse particulates appear to be. Specifically, tephra seems to occur in association with  
352 episodic pulses of particulate matter, often with elevated sulphur levels. The association of a solitary  
353 tephra shard of Katla origin with a sulphur layer contemporary with the 822 eruption of that same  
354 volcano in nearby Iceland serves to emphasise the potential significance of even a single shard in the  
355 ice cores, providing its geochemical composition is reliably distinct and replicable. Affirming previous  
356 work (Jensen et al. 2014; Sun et al. 2014; McConnell et al. 2017; Dunbar et al. 2019), our study  
357 demonstrates that exactly co-registered measurements of distinctive volcanic aerosols (e.g., sulphur)  
358 and fine particle size distribution in combination with exceptionally good age control of polar ice  
359 cores remain the best diagnostic tracer for the presence of tephra within the ice-core strata.

360 At least two of the tephras we have recorded may have an origin in the North Pacific region,  
361 essentially upwind of Greenland given the dominant westerlies. Although only single shards were  
362 analysed for each of these samples, in neither instance do we find evidence that the shards are  
363 weathered. Their association with slightly elevated sulphur levels favours their interpretation as  
364 primary deposits from as yet unidentified eruptions. The sample that yielded the largest number of  
365 analysable shards finds its closest geochemical match with recurrent tephras reported in both  
366 Mexico and mainland Europe; we argue that the geochemical homogeneity shown by these tephras  
367 implies unweathered, primary ashfall, and that the source likely lies in the region around Mexico.

368 It follows that low shard concentration should not negate the potential significance of tephra in the  
369 polar ice cores: unless there are unambiguous signs of physical or chemical weathering or  
370 exceptional levels of coarse terrestrial matter, a single shard can aid the identification of primary

volcanic eruption. Nevertheless, sparse concentrations of tephra present major challenges in terms of ensuring robust geochemical characterisation and the differentiation of potentially altered glass. Where possible, we have obtained multiple analyses on individual grains to gauge the reliability of the geochemical signature and to assess the possibility of glass hydration. We advocate the quantification of tephra-sized particles within ice core samples as a means of evaluating the likelihood of remobilised sediment deposition at the ice core localities. In the absence of firm evidence for the presence of reworked tephra, and considering the exceptional weather conditions needed to transport remobilised material to high altitude polar locations, we propose that tephra encountered in Greenland ice cores potentially relate to primary ash fall, irrespective of low shard concentrations.

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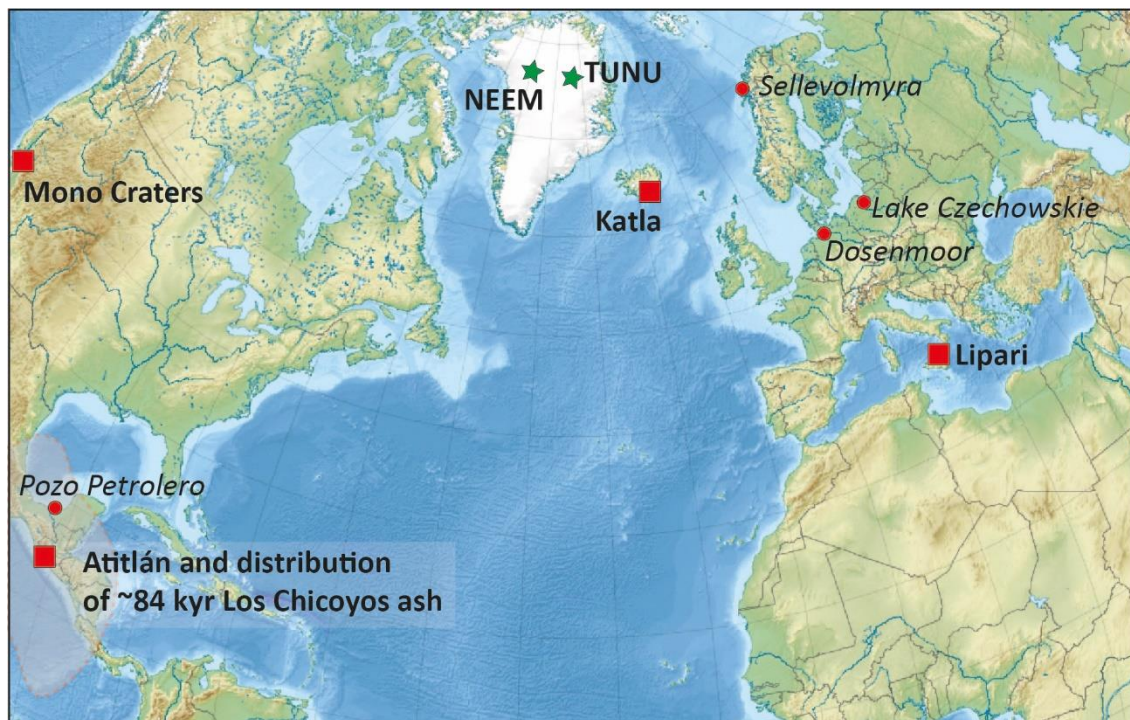
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## Figure captions

**Fig. 1.** Location of NEEM and TUNU coring sites (stars) with respect to volcanic sources (squares) referred to in the text. The locations of sites in which cryptotephra of similar major element geochemical glass composition to QUB-1880 are also indicated (circles; see Plunkett & Pilcher 2018 for a discussion of these tephras).

**Fig. 2.** Positions of tephra samples from the NEEM-2011-S1 and TUNU2013 ice cores in relation to their respective non-sea salt sulphur (nssS) records, and the insoluble particle record from TUNU2013. T or T? indicates the presence of tephra or possible tephra. Examples of non-tephra particles are shown to the right (QUB-1881: charcoal and diatom; QUB-1886: charcoal and fine dust; QUB-1878: fine dust, biogenic silica).

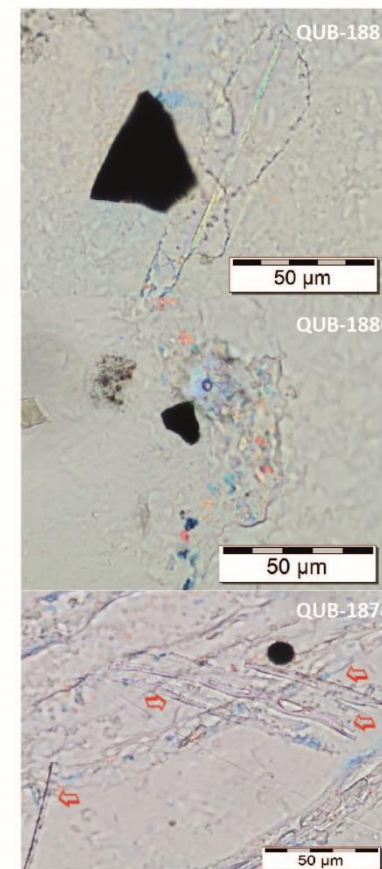
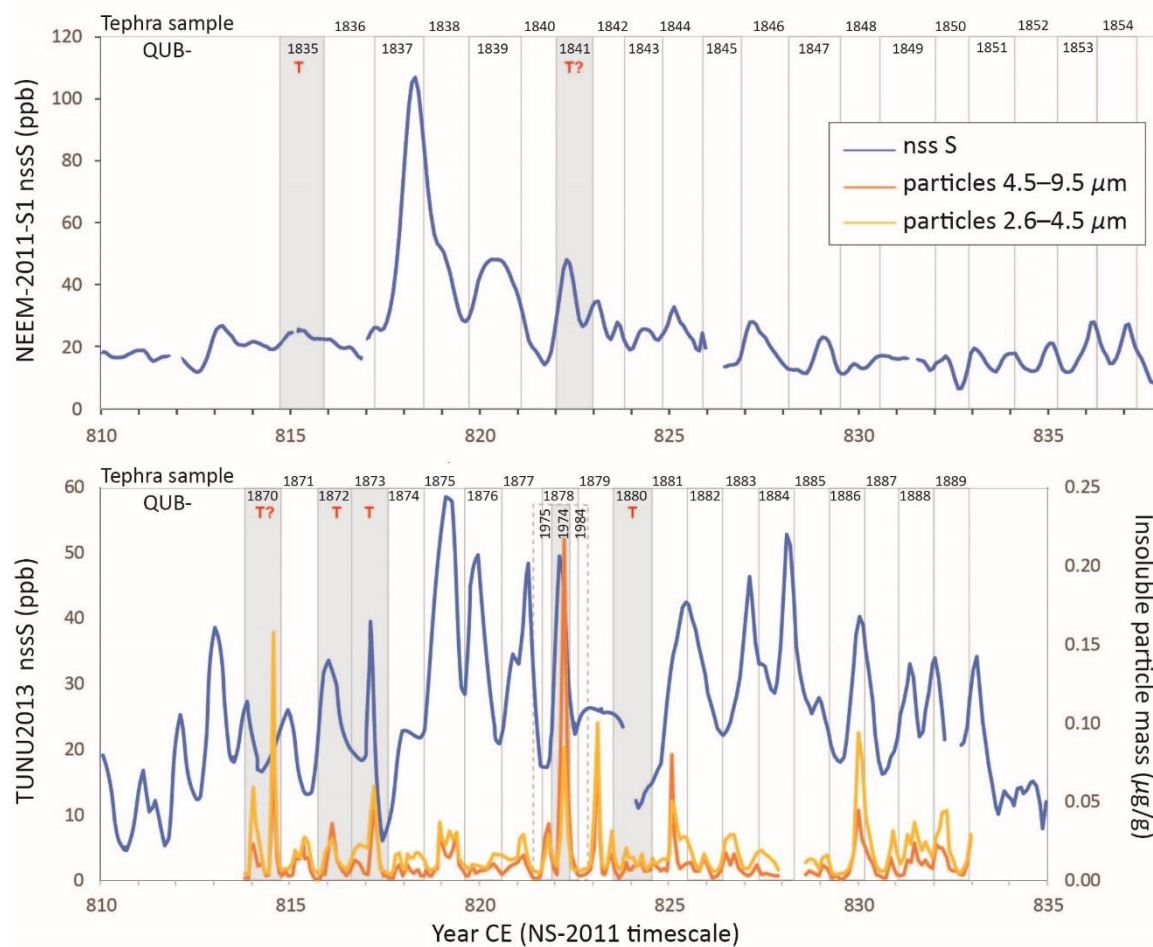


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