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Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality

3rd Nov 23

Dear Ms Gabriel,

I hope you are well?

First please let me apologise for the delay in communicating this decision to you. Your manuscript titled "Long-lasting flare-up of volcanic activity in early medieval Iceland caused climate shocks throughout Europe" has now been seen by 3 reviewers, and we include their comments at the end of this message. They find your work of interest, particularly the multi-disciplinarity, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file. As you revise your manuscript, please also consider the following editorial thresholds:

- Provide compelling evidence for elevated volcanism during a time period previously considered volcanically quiet and discuss critically the resulting impact on climate records.
- Present a clear narrative that links explicitly to the content within the supplementary information; this may require some restructuring of the text.
- Ensure that all raw data and methods are available and reproducible.

Please note that although one of the review reports calls for your raw datasets to be moved to the main manuscript as tables, we are happy for you to keep these within the supplementary information as digital spreadsheets, as long as they are accessible and clearly referenced.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and the completed checklist:

[link redacted]

** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage **

We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

Please do not hesitate to contact us if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

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In particular, the Data availability statement should include:

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Please refer to our data policies at <http://www.nature.com/authors/policies/availability.html>.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Dear editor and authors

Thank you for the opportunity to review this work. In summary, the authors present a very rich dataset, and make interesting and novel observations and discoveries about volcanism in Iceland in the period 700-1000 CE and the associated climate impacts. The scope of the study makes it suitable for publication in this journal. There are many things that the authors do well, but there are also numerous issues, and I recommend revisions before publication. I don't believe additional analysis is necessary but the manuscript would need substantial revision in a few places.

Focus, content, and interpretation:

- I found that the title, the abstract, and the introduction are somewhat misrepresentative of the main focus and content of the study. They claim that the focus of the study is on the so-called Icelandic Active Period (IAP, 751-940 CE), during which there were multiple eruptions. The authors do indeed present some data about several eruptions; however, the lion's share of the results and discussion, and the data-rich figures are on one eruption only, Hrafnkatla, which occurred at the beginning of IAP. Why did the other eruptions within this period did not receive the same level of detailed analysis and discussion as Hrafnkatla? There are other very significant eruptions during this time period, including the Settlement event, and of course, the very large Eldgjá eruption, which is at least as large as Hrafnkatla and maybe even larger. It therefore seems strange (or at least, unexplained) to focus on Hrafnkatla only, especially since the title, abstract and introduction promised to focus on the other eruptions too. I'm sure the authors have reasons for choosing to focus on the one eruption (perhaps the data set for the other eruptions was not as detailed?) but in this case, the title, abstract and introduction should not be overpromising and should be rewritten accordingly. The results about this one eruption are still very interesting and warrant publication.
- One of the most interesting results of the paper is that the new results disagree with previous work about the prevalence of Icelandic volcanism during the studied time period. But this is not explored or explained in the Discussion. Please include a discussion of this and give some possible reasons for the disagreement.
- Figure 5: I didn't understand why the peak values are lower on panel c) compared to b) since the stack is reported to be higher overall in c). Please explain the reasons for this in more detail in the

main text. This seems to be a potentially interesting observation?

Manuscript structure and organisation:

The manuscript is very data-rich and could do with being structured in a better way to help the reader to navigate it.

- Manuscript would benefit greatly from an introduction section on each of the target eruptions. Including but not limited to: size and eruption style; magma composition; which volcanic system they occurred in; tephra size distribution; any information about plume height; length of eruption; etc. Some of this information is included later on, in the Results, but it is required earlier on to be able to better understand the text, and especially the figures.
- There is a large number of supplementary sections (tables, text, figures) and these are not referenced properly in the main text, making it very hard to find the relevant one. Typically the text just says "data is in supplement" with no reference to specific section. Please remedy this.
- Most figures have several panels. Some of the panels are mentioned and discussed in the text; others are not, or only mentioned in passing. Please make sure that all panels are discussed adequately, otherwise it is not clear what they are contributing to the discussion.

Methods and referencing:

- The methods are detailed for most sections (see minor comments on the manuscript file). The section on proximal tephra sampling is, however, missing critical information such as sampling locations, eruption layers which were sampled, references for locations of each, etc.
- Data availability was not satisfactory. In the Methods, it says "all raw data are available in the Supplement" but there was no raw data in the supplement. There were figures, but these are not raw data. Data have to be made available in a spreadsheet or another format where other people can analyse or plot it up. Similarly, it says in the Acknowledgements that raw data will be uploaded to a repository but this had not been done. I appreciate that the authors may want to embargo the raw data but it should be available to the reviewers during the review process.
- Referencing is poor in places, in particular within figure captions. Some of the figures present data from other studies but references are not given.

Figures:

- Generally good but see some comments on the manuscript file

Best wishes,

Dr Evgenia Ilyinskaya

University of Leeds

Reviewer #2 (Remarks to the Author):

The paper focusses on a 150-yr period (751–940 CE) during which there are relatively high levels of non-sea-salt sulphur in Greenland ice cores. Data are presented for various non-sea-salt sulphur peaks to suggest a higher frequency of volcanic activity from Iceland during this time. The paper then attributes the odd climate anomalies experienced in Europe/North Africa in the time period to the long-lived Icelandic Hrafnkatla eruption (751–763 CE).

It is clear from existing Icelandic records that there was more volcanic activity from Iceland between 750 and 940 CE. The new data from the ice cores on this activity is only part of the record, with other studies providing additional key evidence. The new aspect of this study is the link between the volcanism and impact, and this is very speculative. The title of the paper is not justified as the only

impact discussed is related to the Hrafnkatla eruption. Tying the Hrafnkatla episode, with stratospheric sulphur injection only at the end (~763 CE), to the cooling recorded in the European speleothem (Fig. 6d) is tenuous. The cooling recorded commences before 760 CE and so cannot be attributed to the stratospheric injection associated with eruption. In addition, the link to the Nile low stands is also weak as there are other periods when it is low. These linked impacts need to be more critically reviewed and require more discussion.

The paper relies heavily on information presented in both the supplementary information and the supplementary data. The new data, such as the glass chemistry and evidence for the provenance of the tephra needs to be in figures and data tables in the paper. Furthermore, the figure that shows the frequency of eruptions from Iceland, based on the Icelandic database, is key to the main conclusion and should also be in the paper. The paper needs reorganisation to accommodate the data plots and tables.

Comments related to specific sections of the paper and SI:

Line 99 – see comment below on Supplementary Fig. 3.

Line 102 – state the volume of the sample processed.

Line 115 – typically referred to as bulk volume.

Line 188 – this implies that the episode could be related to activity from other systems in addition to Kafa. Evidence needs to be presented or cited if this is the case.

Line 122 – what is the evidence to attribute this to secondary deposition? This requires more detail and discussion.

Line 129: This figure and the data should be in the paper.

Line 133: Reference should be inserted.

Line 137: The new information presented is not enough evidence to indicate that Iceland is highly active and productive in the period. It needs a lot more context before such a conclusion is reached.

The next sentences provide the evidence and are the foundation for this comment so should be moved earlier. Supplementary Fig 6 should be referenced and moved into the paper.

Fig. 1. Would be good to have an inset over Iceland showing all the volcanoes that are known to be active during this period and where samples were taken.

Fig. 2. Hard to distinguish SHET and NHET on (a) as the colours are too similar, and they are not the same as mentioned in the caption, e.g. tropics are grey not black. References for the tephra samples in (b) that are not new (in grey) should be added to the caption. Not sure why F- is in both (c) and (d). The caption should have 'Nc-TI' and 'Nc-Bi' in brackets instead to be consistent with the figure.

Fig. 3. caption for (d): abbreviations in brackets are not the same as those used in the legend on the figure.

Fig. 4. Plot the compositions of the other tephra layers identified, e.g. all the new layers shown in Fig. 2b, and fields for the different Icelandic volcanoes.

Comments on the Supplementary Information:

Line 69, cryptotephra samples: This is missing the detail of what samples were selected, how they were selected, and what length of time a sample represents. It is mentioned elsewhere but it should also be stated here.

Line 98, Sampling: I think this information needs to be in the main paper. It would be good to have annotated photographs or logs. The sampling points should be plotted on a map.

Line 166-167: Sampling strategy for what measurements? In line 99 of the manuscript, it implies that samples were taken "...by exactly co-registered analyses of size resolved insoluble particle

concentrations and sulphur” but on this figure (which is referred to) it seems that sampling was continuous. This needs to be better explained as suggested above (comment for line 69). It would be helpful if there was an additional x-axis with dates so it can easily be related to other figures.

Line 185, S.Fig.2: It is not clear why only fields for those volcanic systems are shown. This needs further justification. I assume because the Iceland record is already well known, e.g., Gudmundsdottir et al., (2016). Other volcanic systems, such as Snaefellsjökull, Askja and Torfajökull, could be shown for comparison. Why are the volcanic fields not marked on S.Fig.4 and S.Fig.5?

Line 204, S.Fig.3: It would be great if images of the other cryptotephra shards could be added.

Line 254, S.Fig.6: This is critical to the argument that Iceland is more active during the highlighted period and thus should go in the main paper.

Line 254, S.Fig.7: This appears the same as Fig. 3 in the paper. It is not clear why it is here.

Line 277, S.Fig.8: Would be good to mark the position of the event on the figure.

Line 295, S.Fig.9: Need to mark on positions of the eruptions.

Supplementary data comments:

Tab 1: This should be a table in the paper. Need to define abbreviations used, e.g., McC18; UniBe (IG). The shard concentrations for T197 and T199 need to be listed if they are Icelandic as it is directly relevant to this study and not be acceptable to just cite an in prep. publication.

Tab 2: This information needs to go as a table in the paper. Define fa-ml, and fa-ca. Need to add photographs or logs of the sections.

Tab 4. Operations conditions: For what? Probably better to include in the paper or SI.

Tab 5. A summary table of the major element compositions should go as a table in the paper.

Tab 6. A summary table of the trace element compositions should go as a table in the paper.

Tab 9. Define MATT. This dataset needs explanation.

Reviewer #3 (Remarks to the Author):

The authors presented a new and interesting idea: An increase in Icelandic volcanism influenced climate conditions in Europe around 700-1000 CE. I think it is a very complex and multidisciplinary paper that combines paleoclimate research, tephrochronology, and glaciochemistry. Probably because of this complexity I found this paper relatively hard to read and follow. Certain parts of the manuscript probably can be reorganized. For example many details from the methods sections could be organized as tables with data and maybe most of the text can be moved to the Supplementary section. I found that certain parts of the research can not be replicated. The authors mentioned R scripts used for statistical correlations of tephra and described statistical methods for background and volcanic peak selections, but they are not provided in the paper.

I think that the information on methods for tephra sample extraction, preparation and measurements is well organized and reported. Probably tephra related plots would benefit with more information about why Icelandic sources are selected. For example, the possible range of major and trace element values for Iceland and other regions in Europe, Kamchatka, Alaska or Japan could be discussed in figure captions.

When I made a plot using (Sigl et al., 2022) data set, I noticed an increase in measured S values in the NEEM-2011-S1 ice core, around the 700-1000 CE interval. I also identified an offset in S levels around 820 C.E. and several increases, for example, around 1450 C.E. or 1700 C.E.. Because there is no

information included on measured uncertainties, it is not clear if these changes are related to measurements or part of the environmental signal. It is specifically required by this journal to access and track all uncertainties. I suggest including uncertainties and also new plots for chemical species (e.g., S, heavy metals and halogens) affiliated with volcanic sources that would show that the 700-1000 CE interval variability is different from other intervals in the last 2000 years. Another potential problem is related to possible changes in chemical species sources or intensities of air mass transport. Such changes are sometimes driven by climate fluctuations impacting atmospheric circulation patterns. In the past, these questions were answered by applying Principal Component Analysis (PCA) to large glaciochemical data sets. A similar test could help to capture changes in volcanic species associated with S and reject possible changes in atmospheric dust transport from the Sahara region that for example has a lot of Fe, Ca and S.

Finally I am not fully convinced that several unusual climate extremes in Europe could qualify as “climate shocks throughout Europe” in the title. History of global volcanism is not well developed yet and we could be missing information on some major explosive volcanic eruptions that are not associated with Iceland. These tropical events, as was demonstrated by the manuscript co-authors in the past, could be responsible for climate extremes during this interval.

In summary, I think this interdisciplinary paper topic is suitable for the journal. I also think the idea to find evidence of a long term increase in Icelandic volcanism around 700-1000 CE in Greenland ice cores deserves proper consideration.

At its present form most of the conclusions are not fully supported by rigorous data analysis that can be replicated by the reader. I would recommend that ice core data will be presented and evaluated for a longer time frame (e.g. the last 2000 years) and it has to be demonstrated statistically, with reported uncertainties, that the interval around 700-1000 CE has unusual patterns in glaciochemistry that exceed variability for major chemical species that are associated with volcanic sources.

I am convinced that the data analysis I suggested is possible and it would reinforce the proposed connections between Icelandic volcanism and increases in some chemical species in Greenland ice cores. I think this alone will start a new chapter in future research that could identify and possibly quantify complex connections between volcanism and climate.

Specific Comments:

Abstract

Volcanic eruptions are generally considered short-lived events, with atmospheric SO₂ emissions limited to a few years.

Comment. Volcanologically it is not a correct statement. Maybe mention that short lived emissions are common for large explosive eruptions.

It would be useful for the authors to define the term “climatic”. Single episodes observed across Europe and north Africa were short extreme weather events and probably will not qualify as changes averaged over 30 years of weather (the authors can define different criteria to help separate climate from weather).

Introduction

It is not just sulfate aerosols (or sulfuric acid aerosols) that are influencing the state of the climate system after volcanic eruptions. It is also tephra and water. It would be useful to define specific types of emissions from volcanoes that the authors are trying to connect with the proposed increase in chemical concentrations measured in Greenland ice cores. They are using Pinatubo as an example

but it is confusing because it was a large explosive caldera forming eruption with a relatively short duration (3-5 months of climatic episode).

It is not clear why Holocene was selected as a target.

“To gain insights into volcanic eruptions and their associated climate impacts over longer time periods (i.e., the last 11,500 years).

I would recommend that the authors include this reference to a seminal work on sulfur species in ice cores: Legrand, Michel R. 1997. “Ice-Core Records of Atmospheric Sulphur.” *Philosophical Transactions of the Royal Society B: Biological Sciences* 352 (1350): 241–50.
<https://doi.org/10.1098/rstb.1997.0019>.

This statement: “Most commonly, non-volcanic background concentrations” is not correct. Only after biogenic and marine sources of sulfate are removed spline or median smoothing is used to evaluate background levels of sulfate.

The references 7 and 12 are not very relevant:

“However, geological records show that this assumption may not hold true, with evidence of eruptions lasting for several decades to centuries 7,12”

A better reference is:

Self, Stephen, Millard F Coffin, Michael R Rampino, and John A Wolff. 2015. “Chapter 24 - Large Igneous Provinces and Flood Basalt Volcanism.” In *The Encyclopedia of Volcanoes (Second Edition)*, edited by Haraldur Sigurdsson, 441–55. Amsterdam: Academic Press.
<http://www.sciencedirect.com/science/article/pii/B9780123859389000249>.

Data sources section

I think the authors did a remarkable job of coalescing different data sources. It would be helpful to the reader if existing tables clearly show ice cores, measured chemical species, and current locations of data. I spent a lot of time trying to find data and understand how the data was synthesized.

1 Long-lasting flare-up of volcanic activity in early medieval Iceland caused climate 2 shocks throughout Europe

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19 20 **Abstract**

21 Volcanic eruptions are generally considered short-lived events, with atmospheric SO₂ emissions limited to a few
22 years. Due to this assumption, state-of-the-art volcanic aerosol forcing reconstructions are biased towards the
23 detection of such events, which has implications for simulating past climate variability with climate models. To detect
24 long-lasting volcanic episodes, we apply a multi-proxy geochemical toolkit on an array of Greenland ice cores across
25 the period 700–1000 CE, a time portrayed as volcanically quiescent, promoting the transition into a Medieval Warm
26 Period. We identify a prolonged episode of volcanic SO₂ emissions (751–940 CE) dominated by Icelandic volcanism,
27 that we term the Icelandic Active Period. This period commences with the Hrafnkatla episode (751–763 CE), which
28 had strong climatic impacts across Europe and north Africa. This study reveals an important contribution of sustained
29 volcanic SO₂ emissions to the pre-industrial atmospheric aerosol burden, offering a natural analogue for solar-
30 radiation modification strategies.

Introduction

The short-lived climate impacts associated with sulfuric aerosols from volcanic eruptions are well established^{1,2} and as a result, volcanoes are considered as one of the closest natural analogues for solar-radiation modification (SRM) strategies which aim to mitigate global warming^{3,4,5,6}. For SRM modelling experiments, the tropical 1991 CE volcanic eruption of Mount Pinatubo is widely used as a reference, largely due it being one of the few eruptions with direct observations of stratospheric aerosols using remote sensing⁷. To gain insights into volcanic eruptions and their associated climate impacts over longer time periods (i.e., the last 11,500 years) we need to exploit polar ice-core records which permit detailed volcanic eruption records to be established via sulfuric acid (H₂SO₄) peaks^{8,9}. However, the detection of volcanic eruptions based on sulfur concentration alone is problematic, as it has several different sources, including marine biogenic emissions, sea-salt spray, and volcanic eruptions¹⁰.

Most commonly, non-volcanic background concentrations of sulfate (or sulfur) in the respective ice-core records are estimated using filters of various time-lengths (e.g., spline, running median). These are then subtracted from total sulfate (or sulfur) concentrations to isolate the volcanic contribution^{2,8,11}.

This approach uses the implicit assumption that volcanic sulfate depositional signals are short-lived (up to three years), due to the short atmospheric lifetime of sulfate¹. However, geological records show that this assumption may not hold true, with evidence of eruptions lasting for several decades to centuries^{7,12}, including eruptions from Iceland^{9,13}. Due to the proximity of Greenland to Iceland, such long lasting eruptions may be imprinted in the ice-core sulfate records, resulting in long-term changes, which introduce ambiguity for secure detection and quantification of volcanic contributions. At present, clear diagnostic tracers unique to volcanic eruptions have yet to be applied within the polar ice core records. Such tracers would facilitate both the detection and the source attribution of volcanic eruptions and thus allow the distinction between both short-lived eruptions and longer lasting volcanic episodes.

Here, we present a set of four distinct diagnostic tracers (cryptotephra, sulfur isotopes, halogens, and heavy metals) within the ice-core records, which when combined, can characterise important source parameters, i.e., the exact location and relative plume height of past eruptions. We apply this multi-proxy toolkit across the time-period 700–1000 CE, a time of marked climatic and societal transitions^{14,15}. This period was thought to be scarce of major volcanic eruptions and experienced minimal perturbations in solar forcing and thus has previously been termed the Medieval Quiet Period (see Figure 2a)¹⁶. Through the application of these diagnostic geochemical proxies, we identify and attribute episodes of long-lasting volcanism to Iceland and consequently revise existing reconstructions of volcanic sulfate deposition which currently underpin all model simulations across this period^{17,18}. Finally, we explore the climate impacts induced by some of these prolonged volcanic eruptions using proxies and documentary evidence.

Results

The seven Greenland ice cores (NEEM-2011-S1, NGRIP1, NGRIP2, EGRIP, RECAP, B19, and TUNU2013) used within this study are located across the Greenland ice sheet (see Figure 1, Supplementary Table 1) between elevations of 2100 m above sea level (a.s.l.) and 3000 m a.s.l. The inset panel in Figure 1 depicts running medians of different time lengths of the NEEM-2011-S1 sulfur concentrations, which are representative of the long-term trends for all ice cores. These ice cores are all dated by annual-layer counting (NEEM-2011-S1, NGRIP1, EGRIP, RECAP, B19, and TUNU2013 records are all volcanically synchronised to the NS1-2011 chronology¹⁹, whilst the NGRIP2 record uses the DRI_NGRIP2 chronology²⁰). Together, they fully cover the period 700–1000 CE, with a dating uncertainty during this 300-year period better than ± 2 years, constrained by two calendar dated cosmogenic age markers in 774 CE and 993 CE (see Methods, Supplementary Table 2). For consistency, we will use the mean of these two age models which may differ by ± 1 year as will be shown later in individual figures. Details on the analytical methods employed for each ice core are provided (Methods, Supplementary Table 1).

We present stacked annual concentration records of multiple volcanic ice-core tracers alongside existing reconstructions of volcanic activity and inferred changes in radiative forcing for the period 700–1000 CE in Figure 2²¹. Stacked non-sea-salt sulfur concentrations ($N = 6$) show an abrupt increase in 751 CE; prior to this, sulfur concentrations were lower overall, with less frequent variability. Median sulfur concentrations between 751 and 940 CE (the latter date marking the Eldgjá fissure eruption from the Katla volcano²²) were elevated by a factor of 1.6 compared to the period 700–750 CE (Figure 2b). Superimposed on these increased background values are frequent sulfate peaks lasting between 1–12 years, with a sulfur concentration increase in 751–763 CE marking this transition. Stacked non-sea-salt chlorine ($N = 6$), fluorine ($N = 2$), and non-crustal bismuth and thallium ($N = 4$) concentration records strongly resemble the overall structure of the non-sea-salt sulfur record, but with a more pronounced step-change at 751 CE (Figure 2c and 2d). Throughout the 751–763 CE episode, element concentrations are elevated (8-fold increase for F⁻, 4-fold increase for Cl, 12-fold increase for Tl, and 5-fold increase for Bi) and remain elevated above pre-event levels for ~ 12 years. Though concentrations decrease again after 765 CE, the overall concentrations remain elevated (3-fold increase for F⁻ and 2-fold increase for Cl, Tl, and Bi) and highly variable in contrast to the first half of the 8th century, where concentrations of non-sea-salt chlorine (2.9 ng/g), fluorine (0.2 ng/g) and the heavy metals thallium (0.04 pg/g) and bismuth (0.06 pg/g) are low. Correlations between volatile metals (Tl, Bi) and volcanic volatiles (Cl, S) are strong throughout the period 751–940 CE, particularly for the 751–763 CE episode, suggesting a common source, transport, and deposition of these species. This is in agreement with Mason et al. (2022)²³, who propose that volatile metals from volcanic sources can be transported to nearby ice-core sites (i.e., 400 km distance) as water soluble aerosols, forming metal-halogen complexes.

To constrain the source of the volcanic events we searched for cryptotephra in the B19, TUNU2013, and RECAP ice cores. Guided by exactly co-registered analyses of size resolved insoluble particle concentrations and sulfur (see Figure 3a, Supplementary Figure 1), we successfully identified cryptotephra in the following years: 753 CE, 757 CE, 763 CE, 766 CE, 781 CE, 877 CE in B19 and TUNU2013. Notably, ice samples correlated to the years 763 CE and 877 CE

102 had high abundances of volcanic glass shards (up to 4000). Through geochemical analyses (see Methods),
103 correlations were made with the Katla, Grímsvötn, Bárðarbunga-Veiðivötn, and Torfajökull Icelandic volcanic systems,
104 as outlined below, with sample identification based on ice core site, depth, and estimated age. A detailed description
105 of shard characteristics and concentrations can be found in the Supplementary data.

106 Major (see Figure 4) and trace element (see Supplementary Figure 2) analysis of individual glass shards from six
107 samples obtained from the three ice cores between 751 and 763 CE can be correlated to the Grímsvötn
108 (B19_149.88m_753CE) and Katla (B19_149.60m_757CE; B19_148.88m_763CE; TUNU2013_158.20m_762CE) volcanic
109 systems. The 763 ± 1 CE glass shards (up to 50 μm in size within all ice samples) were yellowish-brown in colour, with
110 platy morphologies, and a TiO_2 -rich basaltic composition, characteristic of the Katla volcanic system (see
111 Supplementary Figure 3). We geochemically correlated the 763 CE ice-core tephra (B19_148.88m_763CE and
112 TUNU2013_158.20m_762CE, RECAP_297.44m_762CE) with proximal material from the Icelandic Hrafnkatla eruption,
113 including glass from identified tephra horizons in soil sections ~ 20 km from the main Katla vent (see Methods). The
114 Hrafnkatla eruption^{24,25,26}, also referred to as AT-8 or E2 within the literature^{27,28}, was a major tephra-producing
115 eruption from the Katla volcanic system, with an estimated total uncompact volume of $> 4.5 \text{ km}^3$ ²⁸. This is the
116 earliest tephra-producing Katla volcanic eruption during the Common Era to be geochemically identified within the
117 Greenland ice-core records and marks the end of a ~ 12 -year period of continuous volcanic activity from Icelandic
118 volcanic systems which here we term the Hrafnkatla episode.

119 Limited geochemical analyses were obtained for the 766 ± 1 CE sample (B19_148.67m_766CE) and therefore, a
120 definitive source could not be identified. However, the characteristics of this deposit (i.e., the abundance of other
121 material including minerals and black particles of a similar size to the tephra shards) suggest that the shards were
122 deposited at the site via secondary processes²⁹.

123 Analysis of the 781 ± 2 CE sample (B19_147.29m_781CE) provided a correlation to a Grímsvötn tephra identified in
124 Lake Lögurinn, Iceland²⁶ (see Supplementary Figure 4). Within the lake, this horizon has been dated to 788 CE using
125 the lake chronology (constructed using radiocarbon dating and historical tephra). This horizon is thus considered to
126 be the most likely correlative for B19_147.29m_781CE as it is the only Grímsvötn event reported in the proximal
127 records within ± 50 years.

128 A correlation was also made between the 877 CE event in the B19 ice core to the Settlement Layer (877 ± 2 CE) in
129 Iceland using proximal material which was analysed simultaneously (see Supplementary Figure 5). Both the silicic and
130 basaltic components of the event, attributed to the Torfajökull and Bárðarbunga-Veiðivötn systems respectively, were
131 identified in the B19 ice core. The Settlement Layer is widely dispersed across Iceland and is considered an important
132 horizon for separating the pre-historic (i.e., before 877 CE) and historic (i.e., after 877 CE) periods, defining the
133 approximate date of the Viking settlement of Iceland. The silicic and basaltic components of this horizon have
134 previously been identified in the GRIP, GISP2, and TUNU2013 ice cores^{30,31,32}, providing an important tie-point for the
135 synchronisation of records.

The multiple correlations to Icelandic volcanic systems during the 751–940 CE period suggests that these systems were highly active and productive, and thus we term this period the Icelandic Active Period (IAP). This is in agreement with reconstructions of volcanic activity from proximal geological records, which show an increase in activity (i.e., eruption frequency and cumulative tephra volume) of the Katla and Bárðarbunga-Veiðivötn systems during the period 751–940 CE compared to earlier and later time-periods^{24,28} (see Supplementary Figure 6). Centennial mean tephra volume (minimum volume dense rock equivalent (DRE)) from Katla volcanic eruptions was 0.1 km³ between 1–750 CE, increasing to 2.0 km³ between 751–940 CE and decreasing to 0.1 km³ between 941–1755 CE, which volcanologists link to the presence of a relatively shallow magma chamber that was active prior to the 939 CE Eldgjá eruption²⁸.

To further investigate the onset of the IAP, which commenced with the Hrafnkatla episode (751–763 CE), we measured S isotopes in sulfates at high-temporal resolution across this 12-year period on the TUNU2013 ice core (see full details in Methods). Mean $\delta^{34}\text{S}$ values (see Methods for details) across the Hrafnkatla episode are +6.4‰ (see Figure 3b). These values are lower compared to previously measured mean $\delta^{34}\text{S}$ TUNU2013 background samples (+9.5 ‰ $\pm$ 0.93‰)³³, and thus suggest a largely volcanic origin. Background sulfate consists of both marine (21%) and volcanic (~0‰) emissions, and an increase in volcanic sulfate results in lower $\delta^{34}\text{S}$ values within the ice, as observed across the Hrafnkatla episode. Background corrected $\delta^{34}\text{S}$ values over this period are +2.6‰ ($\pm$ 2.5‰) (see Supplementary Figure 7). The anti-correlation between the local S peaks and the $\delta^{34}\text{S}$ further suggests large contributions of sulfur with a volcanic origin. Cryptotephra was identified in samples with low (at or below -0.6‰) $\delta^{34}\text{S}$ values (see Figure 3a).

Previous polar ice-core studies^{33,34,35,36,37,38} have successfully utilised $\Delta^{33}\text{S}$ (see Methods for details) isotopic values to determine whether sulfate aerosol formation occurred in the stratosphere, or more precisely above the ozone layer, which above Iceland is located between 11 and 15 km, thus providing an indication of relative plume height. Our analysis of $\Delta^{33}\text{S}$ across the Hrafnkatla episode has revealed that this volcanic sulfate mainly has values varying between -0.2 and +0.2 ‰ for ~12 years (see Figure 3c). These values are indistinguishable from the 2.s.d. of our non-MIF secondary standard (see Methods) and indicate a ~0 ‰ $\Delta^{33}\text{S}$ value, and hence a dominant tropospheric origin for the S fallout. However, in 762/3 CE, values fall to -0.5 ‰, suggesting that this prolonged elevated tropospheric phase culminated in a short-lived stratospheric phase, which through cryptotephra geochemical analysis we have correlated to the Katla Hrafnkatla volcanic eruption (see Figure 3a). This is the first detection within the ice-core records of anomalous $\Delta^{33}\text{S}$ values for an Icelandic eruption, and unlike tropical stratospheric eruptions (i.e., Samalas 1257 CE and Tambora 1815 CE³³), the $\Delta^{33}\text{S}$ values are much lower for this high-latitude stratospheric Icelandic eruption. This difference can be attributed to the simultaneous deposition of tropospheric and stratospheric S, resulting in a muted anomalous $\Delta^{33}\text{S}$ value.

No sulfur isotope analyses have been performed for the remaining part of the IAP, but sulfur concentrations frequently exceed typical levels which were prevalent for the first half of the 20th century during periods of intensive industrial activity and related sulfur emissions across the North Atlantic region (Figure 5a). Considering the high activity of these Icelandic volcanic systems across the study period, as revealed through the application of the above

proxies, traditional approaches to isolate and quantify volcanic eruptions and their sulfur emissions are inevitably biased. This has resulted in an underestimation in the number, total duration, and SO₂ emission strength of volcanic events. For example, both the reconstructions of Gao et al. (2008)¹¹, the recommended volcanic forcing for the Paleoclimate modelling intercomparison project (PMIP) 3 model experiments, and Plummer et al. (2012)³⁹, did not detect any volcanic events between 751 and 763 CE (see Supplementary Figure 8). This is because existing reconstructions of volcanic sulfate deposition used in these forcing estimates (e.g., NGRIP1³⁹ and NEEM-2011-S1⁴⁰) across the 700–1000 CE period have attributed most of the background variation to non-volcanic sources. Annual mean volcanic sulfate deposition across this period was only 1.0 kg km⁻² yr⁻¹ of sulfate and 2.1 kg km⁻² yr⁻¹ of sulfate for NGRIP1 and NEEM-2011-S1 respectively. We therefore revised existing reconstructions of volcanic sulfate deposition, now attributing long-term background variations of sulfur as volcanic in origin (see Methods). As a result, we were able to reconstruct overall higher volcanic sulfate deposition rates for NGRIP1 (2.4 kg km⁻² yr⁻¹ of sulfate) and NEEM-2011-S1 (4.1 kg km⁻² yr⁻¹ of sulfate) (see Figure 5). Additionally, we reconstruct new volcanic sulfate depositional records from NGRIP2, B19, TUNU2013, and EGRIP, thus doubling the number of ice-core sulfate records from Greenland currently used in reconstructions of volcanic forcing (i.e., by PMIP4) during the Medieval period^{18,21}.

Differences between the analysed ice cores, with the same events not detected in all ice cores in some instances, may mostly be attributed to the location and elevations of the drill sites (e.g., TUNU2013 and B19 are 700–800 m lower than EGRIP and NGRIP) as these will subject the sites to both different atmospheric circulation and different emission sensitivities towards major volcanic source regions.

Discussion

Previous assumptions used in volcanic forcing reconstructions, where volcanic eruptions are considered as short-lived stochastic events, are not supported across the period 700–1000 CE. Our application of a multi-proxy geochemical toolkit has revealed heightened volcanic activity from several Icelandic volcanic systems and includes both long-lasting, multi-centennial episodes such as the Icelandic Active Period (IAP) (751–940 CE) and short-lived episodes such as the Hrafnkatla episode (751–763 CE). Both are characterised by prolonged and elevated enrichments of volcanic metals (Tl, Bi) and volatiles (F, Cl, S).

Within Antarctic ice cores, a pronounced elevated halogen and metal enrichment **lasting 192 years** was attributed to regional West Antarctic volcanism^{12,23,41}. Mason et al. (2022)²³ suggested that the long-lasting excessive deposition of these volatile species is indicative of a proximal eruption, with magma source chemistry, eruptive processes and plume transport also contributing to the signature within the ice. Whilst a great deal is still unknown about the behaviour of many volatiles during transport, prolonged stratospheric transport (i.e., from a distal eruption) is likely to result in greater loss due to scavenging, and thus reduced or negligible deposition on polar ice sheets. Modern plume and ground-based measurements of the Icelandic Holuhraun 2014–2015 CE volcanic eruption support this, with **Ilyinskaya et al. (2017)**⁴² observing a decrease in atmospheric burdens of HF, HCl, and volatile metals with distance from the source. Owing to this decay, and the proximity of Iceland to Greenland (~1800 km), we suggest that

207 in the Greenland ice cores this metal signature could be used as an indicator for Icelandic eruptions in the absence of
 208 cryptotephra. These trace metals are now routinely analysed over the entire length of deep ice cores and thus are
 209 readily available for improved source attribution of volcanic signatures within ice cores ^{12,20}.

210 We propose the prolonged elevated concentrations of volcanic volatiles and volatile metals observed across the IAP
 211 can best be explained by continuous volcanic activity dominated by several Icelandic sources (Katla, Bárðarbunga-
 212 Veiðivötn, and Grímsvötn). This is supported by the general increased eruption frequency from these systems (see
 213 Supplementary Figure 6), with Katla producing 10 times more tephra volume between 750 and 950 CE compared to
 214 time-periods before and after ²⁸. We postulate that eruptions across the IAP encompassed a wide range of volcanic
 215 eruption types, involving explosive stratospheric eruptions as well as prolonged fissure and lava flood eruptions and
 216 quiescent degassing. This includes, for example, the long-lasting Hallmundarhraun lava flow, which occurred between
 217 880 and 910 CE, and is considered the third largest effusive eruption since the settlement of Iceland (~877 CE) with
 218 an estimated erupted volume of up to 8.5 km³ ^{43,44}.

219 Overall, and supported by the sulfur isotopes, a large fraction of the aerosol emissions remained predominantly in
 220 the troposphere/lowermost stratosphere (LMS). This strong and long-lasting, tropospheric S contribution led to the
 221 estimate of volcanic sulfate deposition in Greenland across this 300-year period being revised and increased by a
 222 factor of 2 (see Figure 5b and 5c), and thereby challenges the notion of this being a Medieval Quiet Period. This has
 223 important implications for climate model simulations, such as those from PMIP3 and PMIP4, which use the year 850
 224 CE as the start of the *past1000* simulations covering the pre-industrial millennium (850 to 1849 CE) ^{17,18}. These
 225 models use only stratospheric volcanic forcing where the tropospheric aerosols are not included, in contrast to
 226 historic climate simulations (starting in 1850 CE), which also include tropospheric aerosol forcing.

227 The role of tropospheric volcanic aerosols in Earth's system should not be neglected. These aerosols not only have
 228 direct impacts (i.e., reflection of incoming radiation and surface cooling ⁴⁵), but can act as ice-nucleating particles
 229 (INPs) and cloud condensation nuclei (CCN), thereby influencing cloud properties and the hydrological cycle ⁴⁶.
 230 Despite the importance of tropospheric aerosols, little is known about pre-industrial concentrations, a time when the
 231 atmosphere showed greater sensitivity towards tropospheric volcanic aerosol inputs (e.g., from quiescent degassing)
 232 due to the absence of anthropogenic aerosols ^{47,48}. Developing a baseline of pre-industrial sulfuric aerosol
 233 concentrations would therefore help constrain uncertainties in radiative forcing estimates which are used in climate
 234 model simulations.

235 These ongoing uncertainties and different aerosol baseline conditions during the pre-industrial era have presented
 236 challenges when assessing the climate impacts of prolonged volcanic episodes. To investigate the onset of the IAP,
 237 which commences with the Hrafnkatla episode, we compare forcing reconstructions to palaeoenvironmental records
 238 from the North Atlantic region where they are abundant and well-dated. Due to atmospheric circulation patterns,
 239 which confine aerosols from high latitude volcanic eruptions to their respective hemispheres, the climatic impacts of
 240 large Icelandic eruptions are more spatially limited than those of tropical eruptions such as the 1991 CE eruption of
 241 Mount Pinatubo. The climatic impacts associated with the Hrafnkatla event are generally comparable to other large

Icelandic volcanic eruptions such as Eldgjá 939 CE and Laki 1783 CE, with strong winter cooling anomalies across Europe (i.e., Oman et al. 2006⁴⁹; Zambri et al. 2019⁵⁰) and perturbations to the hydrological cycle^{49,51,52,53}. At around 762–763 CE, a pronounced cooling anomaly (up to 1°C) is recorded in two speleothem records from central Europe (Milandre cave, Switzerland⁵⁴ and Spannagel cave, Austria^{55,56}) (see Figure 6d). Historical records cite the winters of the same and subsequent years (762–764 CE) among the harshest of the Common Era, with widespread frost and ice, crop failures, and famines documented from Ireland to the Black Sea^{57,58,59} (see Figure 1). Indeed, the first-hand eyewitness account of Theophanes the Confessor documents an extremely severe winter in the area of the Black Sea and the Bosphorus from October 763 CE to February 764 CE and describes that “in early October [763 CE], there was very bitter cold, not only in our land, but even more so to the east, the north, and the west, so that on the north coast of the Pontos [the Black Sea] to a distance of 100 miles the sea froze [...] In the month of February [764 CE] this ice was [...] split up into many different mountain-like sections. [...] One of the icebergs struck the jetty of the Acropolis [of Constantinople] and crushed it”^{60,61,62}. These harsh conditions documented across Europe coincide with a period of extreme annual low stand values of the river Nile (20 of the 52 lowest 5th percentiles occurred between 750 to 790 CE)⁶³ (Figure 6c). The flow of the river Nile is highly sensitive to shifts in the Intertropical Convergence Zone (ITCZ), with the headwaters of the Nile located in the equatorial regions. The ITCZ is displaced southward in response to the high atmospheric burden of sulfate aerosols and subsequent cooling within the Northern Hemisphere atmosphere following high-latitude eruptions, resulting in varied precipitation patterns in the equatorial regions and reduced stream flow of the Nile^{49,64,65}.

In contrast to many historic large eruptions, no major summer cooling anomaly is recorded in Northern Hemisphere tree-ring reconstructions in the year following the 763 CE Hrafnkatla eruption (i.e., Schneider et al. 2015⁶⁶; Büntgen et al. 2020⁶⁷). The cause for this absence of a summer cooling anomaly is unknown; however, given that the explosive Hrafnkatla event followed a 12-year period of sustained aerosol emissions, increased background aerosol concentrations may have modified the climatic response. Arctic sea-ice plays an important role in amplifying and sustaining climatic responses to large explosive volcanic eruptions, and typically, following an explosive volcanic eruption there is an expansion in sea-ice^{68,69,70}. In contrast, methanesulphonic acid (MSA) concentrations (a proxy for sea-ice) from the TUNU2013 Greenland ice core (Figure 6b) show no clear indication of a strong sea-ice expansion around the 760s CE. They do however, indicate a general reduction in Arctic sea-ice across the IAP, coincidental with an increase in surface air temperature, peaking in the late 10th century (Figure 6a). This transition into a North Atlantic Medieval Warm Period (MWP) coincided with the Viking expansion across the Arctic region to settle first in Iceland (~877 CE) and then in southern Greenland (~985 CE)^{71,72}. Arguably, these warmer temperatures and ice-free conditions may have facilitated such an expansion. Ultimately, this is an area that requires further research, with continued uncertainties regarding the climatic response and feedbacks associated with prolonged volcanic episodes, particularly during the pre-industrial era. Further insights could not only allow us to better understand these societal and climatic transitions but may have important implications for SRM studies aiming to mitigate global warming, particularly in the Arctic.

The aerosol clouds generated by large explosive eruptions are generally short-lived, which contrasts with the permanent aerosol cloud that would be generated during SRM^{1,3,4}. As a result, Robock et al. (2013)⁷³ argue that the observed climate response using this natural analogue would be different, particularly on longer time scales. Prolonged volcanic episodes such as the IAP, however, would offer an alternative volcanic analogue, with the continuous elevated sulfur concentrations across this 190-year period more closely representing the deployment of SRM in the high-latitudes. Therefore, more research into these prolonged volcanic episodes may provide the necessary information to robustly assess both the effectiveness and uncertainties regarding SRM, particularly the ramifications on the biosphere, cloud properties, and precipitation patterns.

In conclusion, our application of a multi-proxy geochemical toolkit has revealed that the period 751–940 CE was a time of heightened Icelandic volcanic activity, which commenced with the Hrafnkatla episode in 751 CE. This period was characterised by heavy metal and halogen enrichments, with these species most pronounced at the time of local Icelandic eruptions. We have therefore postulated that both heavy metals and halogens can be used as distinct tracers of Icelandic volcanic eruptions within the Greenland ice core records and aid in discerning the source in the absence of cryptotephra. Through revising volcanic sulfur deposition across this period, we identified a greater tropospheric/LMS contribution of sulfate aerosols, which is currently not considered in existing volcanic forcing estimates. We found that the explosive stratospheric end-phase of the Hrafnkatla episode coincided with a strong winter cooling anomaly throughout Europe and could be considered as a likely cause. Unlike other large explosive volcanic eruptions however, no major summer temperature anomaly has been observed thus far. We suggest that more research into prolonged volcanic episodes, such as the IAP, is required due to the uncertainties in the associated climate feedbacks to sustained tropospheric aerosol emissions, especially during the pre-industrial period. This is particularly important for the last deglaciation to early Holocene, during which time such long-lasting episodes occurred frequently, with some lasting up to 70 years^{9,74}. Finally, these prolonged volcanic episodes more greatly resemble potential deployment of SRM in the Arctic, in contrast to short-lived volcanic eruptions. Therefore, enhancing our understanding of the climate response to these prolonged volcanic episodes may facilitate their use as better analogues for SRM and its wider impacts.

Methods

Ice-core analysis. Information regarding the seven Greenland ice-core records used within this study (NEEM-2011-S1, NGRIP1, NGRIP2, EGRIP, RECAP, B19, and TUNU2013) can be found in Supplementary Table 1. Elemental data (including sulfur, chlorine, bismuth, and thallium) for NGRIP2, NEEM-2011-S1, and TUNU2013^{40,19,20} ice cores were utilised alongside new measurements of these elements in the B19 ice core (78.00°N 36.40°E, 150.4 m depth, 2234 m a.s.l.)⁷⁵. All data were obtained through continuous flow analysis (CFA) coupled to two inductively coupled plasma mass spectrometers (ICP-MS, Element2, Thermo Scientific) at the Desert Research Institute, USA (DRI), providing a simultaneous determination of a broad spectrum of elements at a sub-annual resolution¹². Additional sulfate (SO₄²⁻), chloride (Cl⁻), and fluoride (F⁻) measurements for EGRIP and NGRIP1 were undertaken at a 5 cm resolution at the

Alfred Wegner Institute, Germany (AWI), and Physics of Ice, Climate and Earth, Denmark (PICE), respectively, using ion chromatography. For the determination of anionic species in EGRIP, a two-channel Dionex ICS 5000+ reagent-free HPIC system (Thermo Fisher) with a Dionex IonPac AS18-Fast-4 μm analytical column was used. Details of the entire sampling procedure and analytical conditions are described in detail in Schrod et al. (2020)⁷⁶. Between 700 and 1000 CE the time resolution is about 0.5 years in EGRIP. For quantification of ionic species in NGRIP1 a two-channel Dionex 500 microbore ion chromatograph was used^{39,77,78} resulting in a time resolution of about four samples per year during the first millennium CE. In addition, methanesulphonic acid (MSA) was measured at DRI continuously over the TUNU2013 ice core using an electrospray triple-quad mass spectrometer (ESI-MS-MS)⁷⁹ alongside stable water isotope ratios ($\delta^{18}\text{O}$), determined using a cavity ring-down water isotope analyser (L2130-i, Picarro Inc)⁸⁰. All parameters measured for each ice core and used within this study can be found in Supplementary Table 1.

Ice-core data processing. We estimated non-sea-salt sulfur (nss-S) concentrations by correcting total S concentrations for the sulfur contribution from sea salt using co-registered Na concentrations as a sea salt tracer and assuming a sulfur to sodium mass ratio in bulk sea water of 0.084⁸¹. Similarly, we estimated non-sea-salt chlorine (nss-Cl) concentrations by correcting total Cl concentrations for the chlorine contribution from sea salt using co-registered Na concentrations as a sea salt tracer and assuming a chlorine to sodium mass ratio in bulk sea water of 1.796⁸². To calculate the non-crustal fractions (nc-Tl and nc-Bi) and enrichment factors (Tl-EFCe and Bi-EFCe, see Supplementary Figure 9), we used cerium (Ce) as a conservative tracer for mineral dust and the “mean continental crust” Tl/Ce ratio of 0.01172 and Bi/Ce of 0.00198, respectively⁸³. Ce concentrations were scaled by a factor of 1.7 to account for an under-recovery of Ce (of 60%) during the continuous-flow analysis at the Desert Research Institute, whereas recovery was ~100% for both Tl and Bi⁸⁴.

Targeted sampling. CFA measurements of insoluble particles at two size fractions (2.6–4.5 μm and 4.5–9.5 μm) were used alongside peaks in sulfur to assist with isolating events and sampling for tephra. Particle measurements were undertaken using an inline Abakus® laser particle counter⁸⁵ during the CFA analyses at DRI. Although the particle concentrations alone cannot be considered as a direct proxy for tephra, the application of this approach, based on the exact stratigraphic co-registration of insoluble particulates and volcanic volatiles in the polar ice cores, has proven highly successful for detecting cryptotephra layers^{29,32,86,87,88,89,90,91}. One 11 cm-long sample was obtained for tephra analysis from the RECAP ice core between 297.44–297.55 m for which a non-targeted continuous sampling approach was followed as described by Cook et al.⁹².

Ice-core chronologies. The NS1-2011¹⁹ and DRI_NGRIP2²⁰ chronologies are used within this study. Both chronologies were constructed through annual layer counting of seasonally varying impurity tracers constrained by well-dated stratigraphic markers such as historic volcanic eruptions, volcanic dust veils, and solar energetic particle events (see Supplementary Table 2 and Table 3). Dating uncertainty in both chronologies during the period 700–1000 CE is ± 2 years. This is supported by the occurrence of short-lived cosmogenic nuclide events in 774 CE and 993 CE, which are observed in both the ice core and tree-ring records^{19,93,94} and by exact matches of dendrochronologically dated volcanic eruptions and corresponding ice-core tephra identifications in 822/823 CE^{29,93} and 946/947 CE²². Both chronologies are within errors consistent with the GICC21 chronology⁹⁵, but are more precise during the period 700–

1000 CE because of the additional age constraints. To transfer the age of the RECAP ice core tephra sample (available on the GICC05 chronology⁹⁶) to the NS1-2011 chronology, we used an age transfer of +7 years following Sigl et al.¹⁹.

Cryptotephra analysis. Targeted tephra samples were obtained for 11 volcanic events from TUNU2013, B19, and RECAP ice cores and analysed at Queen's University Belfast, UK (QUB); University of Bern, Switzerland (UniBe); and the University of Edinburgh, UK (Edin), respectively (see Supplementary data). The targeted sampling approach allowed us to sample comparably large cross sections of archived ice ranging between 16.5 cm² (RECAP) and 15 cm² (B19) thus increasing the chance of retrieving greater abundances of tephra shards. In addition, samples that were analysed for S isotopes were also centrifuged prior to analysis, facilitating the screening for cryptotephra. The method for the preparation for tephra identification and geochemical analysis of samples analysed at QUB and Edin can be found in Abbott et al. (2021)⁸⁶ and Cook et al. (2018)⁹² respectively. The preparation of samples for tephra identification and geochemical analysis at UniBe followed a method adapted from several recent papers^{97,98,99,100} and the full methodology used within this study can be found in the Supplementary Information. Samples where glass tephra shards were identified were analysed using Electron probe microanalysis (EPMA) at the Institute of Geological Sciences, University of Bern, Switzerland. The samples were carbon coated prior to major elemental analysis using a JEOL JXA-8200 probe, with operating conditions of an accelerating voltage of 15 kV, beam current of 2 nA and beam diameter of 5 µm. Secondary standards of Cannetto Lami Lava, Lipari and BCR-2Ga were measured during the analytical session to ensure stability and allow session comparisons. Full details about the operating conditions and secondary standards can be found in the Supplementary data.

Following the filtering of all obtained data to totals > 90wt% and the removal of Cl and P₂O₅ (to facilitate correlation with other data sets), the data were normalised to an anhydrous basis following standardised practices¹⁰¹. The data were further inspected to remove any outliers, both those occurring from analytical issues (e.g., analysis of the resin) and those which appeared outside of the main population. All raw data can be found in the Supplementary data. Data were then correlated to published tephra geochemical data, with potential correlations explored graphically using the statistical software R (version 1.4.1717). In order to make robust correlations, samples were analysed alongside proximal material when available, which was obtained in summer 2021/2022 fieldwork. The full preparation method for proximal material and sampling strategy can be found in the Supplementary Information.

Trace element analysis was conducted on individual glass shards from four ice core and three proximal samples covering the period 751–763 CE using laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) at St Andrews Isotope Geochemistry lab, UK (StAiG). Laser ablation used an Applied Spectra RESOLUTION system, featuring a Laurin Technic S155 2-volume cell under a pure He atmosphere (370 mL min⁻¹) with supplemental N₂ (5 mL min⁻¹) to improve sensitivity in the ICP-MS. Analyses were undertaken using a 20 µm beam diameter and a laser energy of 3.5 J cm⁻² with a 4 Hz repetition rate, 20-s background measurement, and 30-s ablation time. The laser cell gases and ablated sample were mixed with Ar (1 L min⁻¹) before being homogenised in a signal smoothing device prior to introduction to an Agilent 8900-QQQ for mass spectrometry. Measurements of glass standards of Old Crow, Sheep Track, and ML3B, and synthetic glass standards of NIST610 and T1G which were used to correct for drift, bracketing measurements, can be found in the Supplementary data. Samples were normalised using Al₂₇, which was determined

from the same samples during previous EPMA at the UniBe. A detailed methodology can be found in Pearce et al.¹⁰². All raw data can be found in the Supplementary data.

Sulfur isotope analysis. The ratio of isotopic ³⁴S, ³²S, can be used to determine the origin of the sulfate (i.e., marine or volcanic)^{103,104,105}. This distinction can be made through the measurement of $\delta^{34}\text{S}$ (defined as $\delta^x\text{S} = ((^x\text{S}/^{32}\text{S})_{\text{sample}} / (^x\text{S}/^{32}\text{S})_{\text{reference}}) - 1$, where x is 33 or 34). Marine sulfate typically has values between 16–20 ‰, whilst volcanic isotopic values are closer to zero^{105,106,107,108,109}.

By measuring the minor isotope, ³³S, one can assess Mass-independent Fractionation (MIF) by calculating $\Delta^{33}\text{S}$ (defined as $\Delta^{33}\text{S} = \delta^{33}\text{S} - ((\delta^{34}\text{S} + 1)^{0.515} - 1)$). $\Delta^{33}\text{S}$ isotopic values can be used to determine whether sulfate aerosol formation occurred in the stratosphere (i.e., above the ozone layer) since UV photochemical reactions generate MIF and non-zero $\Delta^{33}\text{S}$ ^{35,110,111,112}. As a result, sulfur deposited over the ice sheet that has reached the stratosphere has $\Delta^{33}\text{S} \neq 0$. Due to the ozone layer, which filters out radiation of the specific wavelength that causes MIF, the sulfate which remains in the troposphere/LMS is not exposed to UV radiation. Therefore, Mass-Dependent Fractionation (MDF) is generated and tropospheric/LMS sulfate has a $\Delta^{33}\text{S}$ isotopic value that is equal to zero. Through these different fractionation processes, it is possible to distinguish stratospheric sulfate from tropospheric/LMS sulfate and provide an insight into the explosivity and plausible plume heights of past eruptions.

We undertook S isotopic measurements on samples from TUNU2013 across an elevated period of sulfate concentrations. TUNU2013 was continuously sampled at a 3 cm depth resolution (cross sections of 7 cm²) between 740 and 765 CE for sulfur isotope analysis at the University of St Andrews Isotope Geochemistry Laboratory (StAiG), UK. The detailed methodology can be found in the methodology of Burke et al.³³. Briefly, the sulfate concentration of each sample was first determined by ion chromatography using a Metrohm 930 Compact IC Flex. Sulfate was then extracted from the sample using column chemistry and triple sulfur isotopes (³²S, ³³S, and ³⁴S) were subsequently measured using a Neptune Plus Multi-Collector-ICPMS¹¹³. These measurements were conducted alongside an established secondary standard (Switzer Falls¹¹⁴) and procedural blanks. The procedural blank values (0.18 ± 0.08 nmol S, $\delta^{34}\text{S} = 6.8 \pm 4.3$ ‰, 1 s.d., $n = 11$) were used to correct all measured ratios. The $\delta^{33}\text{S}$ and $\delta^{34}\text{S}$ values were used to calculate $\Delta^{33}\text{S}$. The external reproducibility of the secondary standards run alongside these samples (0.19‰, 0.08‰, and 0.09‰ for $\delta^{34}\text{S}$, $\delta^{33}\text{S}$ and $\Delta^{33}\text{S}$ respectively, 1 s.d., $n = 9$) is used as the reported uncertainty on isotope ratios. For some samples there was only enough sulfate for a single isotope measurement, and thus the uncertainty on these samples is slightly elevated and was calculated from the external reproducibility of the secondary standard when only one replicate was considered (0.22‰, 0.1‰, and 0.1‰ for $\delta^{34}\text{S}$, $\delta^{33}\text{S}$ and $\Delta^{33}\text{S}$ respectively, 1 s.d., $n = 9$). The isotope data can be found in the Supplementary data. Following the approach of Baroni et al.³⁸; Gautier et al.¹¹⁵,

$$\delta^x\text{S}_{\text{volc}} = \frac{(\delta^x\text{S}_{\text{meas}} - f_{\text{background}} \delta^x\text{S}_{\text{background}})}{f_{\text{volc}}}$$

we use the mean sulfate concentration and sulfur isotope composition from TUNU2013 non-volcanic background samples analysed throughout the common era to solve for the sulfur isotopic composition of volcanic sulfate (see Supplementary Figure 7). Uncertainties on this calculation are propagated using Monte Carlo simulations, and only samples with more than 57% volcanic sulfate are plotted, similar to that suggested by Gautier et al.¹¹⁵.

Volcano detection. Several statistical methods have been applied in previous studies to the ice-core sulfate records to isolate volcanic events from the background signal^{116,117}. These methods often use running medians of multi-decadal lengths for the separation of volcanic and non-volcanic sulfate. Their application has proven successful in the detection of climate impacting (medium and large sized) volcanic events, but ultimately has resulted in an underestimation in the total amount of volcanic sulfate. Within this study, we therefore adjusted the previous methods^{40,116,117}, across the NEEM-2011-S1, NGRIP1, NGRIP2, EGRIP, B19, and TUNU2013 ice core records, as follows: [1] a 91-year Running Median (RM) was used on the annually averaged nss-S record on periods unaffected by strong changes in volcanic background emissions (i.e., 300–700 CE for all cores but B19, for which 950–1250 CE was used) to estimate the natural background sulfate levels; [2] a Median of Absolute Deviation (MAD), calculated from the RM, was used in peak detection. As with the calculation of the RM, the MAD for the period before (i.e., 300–700 CE and 950–1250 CE for B19) was used. A peak was considered volcanic if it passed the upper threshold ($K=3$, estimated as $RM + 3 * MAD$). The duration of the event was determined when it passed the lower threshold ($K=1$, estimated as the $RM + 1 * MAD$). These upper and lower thresholds were selected by validation on well-known historic eruptions; [3] volcanic peaks were then removed to calculate the non-volcanic background (S RRMi). To further calculate the amount of sulfate deposited, S RRMi (from 300–700 CE and 950–1250 CE for B19) was subtracted from the average annual nss-S and then multiplied by the accumulation rate of the drill site; [4] finally, volcanic flux was calculated for each event by summing the sulfate deposited across the total duration of the event.

Temperature reconstruction for Milandre cave (Switzerland) 600–1200 CE. The Milandre cave (see Figure 1) temperature reconstruction is based on oxygen isotope measurements ($\delta^{18}O$) of speleothem calcite and the data can be found in the Supplementary data. The statistically significant correlation ($r^2=0.56$, $p=0.0001$) between stalagmite calcite $\delta^{18}O$ ⁵⁴ and historical and instrumental temperature records from western Switzerland¹¹⁸ allows the development of a transfer function to calculate mean annual air temperatures for the last two millennia for western Switzerland^{119,120}.

Temperature reconstructions from Greenland $\delta^{18}O$ stack 600–1120 CE. As a proxy for annual mean surface air temperatures over Greenland we collated published stable oxygen isotope ($\delta^{18}O$) records across Greenland fulfilling the selection criteria: [1] continuous coverage between 600 and 1120 CE, [2] with annual time resolution, and [3] dating precision and accuracy better than ± 3 years. A total of six records met these requirements, from the Dye-3, NGRIP, NEEM, GRIP, EGRIP and TUNU2013 ice cores^{95,121,122}. We then compiled our dataset of these individual $\delta^{18}O$ anomaly records (relative to a 1901–1990 reference interval) into a single stack by calculating the arithmetic mean $\delta^{18}O$ anomaly for each year. We further apply a 31-year running mean filter to our time series and estimated temperature anomalies, by applying the spatial calibration slope for Greenland of $10.67^\circ C$ per ‰^{123,124}.

Author contributions. I.G. and M.S. wrote the original manuscript and constructed figures. I.G., P.M.A., G.P., and E.C. undertook tephra major element geochemical analyses. I.G. and J.T.S. undertook trace element geochemical analyses. M.S. and J.R.M. contributed ice-core glaciochemical records. J.R.M. contributed ice core samples for subsequent

measurements of tephra, S isotope analysis. J.R.M. and N.C. drilled and undertook continuous measurements of ice cores with DRI. A.B., W.H., and P.S. analysed S isotopes. D.F. contributed Milandre cave temperature reconstruction. M.H. and M.B. undertook sulfate, fluoride, and chloride analyses for the EGRIP ice core. B.A.O. contributed proximal information regarding Icelandic volcanic activity during the study time period. J.K.P. contributed historical documentary records. All authors contributed to improving and editing the paper.

Acknowledgements. I.G., P.M.A, and M.S. received funding from the European Research Council under the European Union's Horizon 2020 research and innovation programme (grant agreement no. 820047). M.S. received funding from the Geoscience Department and the NFR TOPPFORK project "VIKINGS" (grant no. 275191) at the University of Oslo. I.G. received funding from the Swiss Polar Institute and BNP Paribas Swiss Foundation. Tephra research by E.C. was funded by the ERC project 'ice2ice' (grant agreement 610055). W.H. and P.S. are funded by a UKRI Future Leaders Fellowship (MR/S033505/1). This research benefited from the participation of the authors in the Volcanic Impacts on Climate and Society (VICS) working group of the Past Global Changes (PAGES) project. This work uses data from the NGRIP, EGRIP, and RECAP ice core projects directed and organised by the Physics of Ice, Climate and Earth, at the Niels Bohr Institute, Denmark. The authors acknowledge the scientific consortia and funding agencies associated with each ice core project. The collection and analysis of the TUNU2013 and NEEM-2011-S1 ice cores were supported by US National Science Foundation grants (no. 1204176, 909541) to J.R.M. The B19 ice core was obtained as part of the North Greenland Traverse project, supported by the GRIP ice core drilling project. Analysis of the B19 ice core was funded by the Desert Research Institute, USA. M.S. and P.M.A. thank the Alfred Wegner Institute, Germany and the NSF-Ice core facility, USA for their hospitality and assistance when sampling the TUNU2013 and B19 ice cores. Thanks to Sepp Kipfstuhl for providing access to the B19 ice core. Thanks to Pierre Lanari, Coralie Vesin, and Hugo Dominguez Carranza for help with EPMA analysis at the University of Bern. Thanks to Thor Thordarson for providing detailed information regarding the Hallmundarhraun lava field and support and guidance during fieldwork in Iceland. Thanks to Esther Gudmundsdóttir for providing Lake Lögurinn data for geochemical correlations. Thanks to Francis Ludlow for discussing documentary sources from Europe.

Competing Interests. All authors declare no competing interests.

Data Availability. NEEM-2011-S1 ice core data is available at PANGAEA¹³⁸ and at the Arctic Data Center¹³⁹. TUNU2013 and NGRIP2 ice-core data is available at the Arctic Data Center^{140,141,142}. NGRIP1 ice core data is available at PANGAEA¹⁴³ and from the Physics of Ice, Climate and Earth, University of Copenhagen: https://www.iceandclimate.nbi.ku.dk/data/2012-12-03_NGRIP_SO4_5cm_Plummet_et_al_CP_2012.txt. New ice-core data from these cores (e.g., heavy metals) and from the B19 and EastGRIP ice cores as well as all geochemistry data (S-isotopes, cryptotephra) will be uploaded to PANGAEA: doi: XXX. Volcanic sulfur injections are available at the World Data Center for Climate (WDCC) at DKRZ¹⁴⁴. Nile River flow data was obtained here: <https://doi.org/10.1029/2004GL022156>⁶³. Spannagel cave data is available at NOAA¹⁴⁵ and Milandre cave data will be made available upon publication of the full-length record (Fleitmann et al.¹²⁰).

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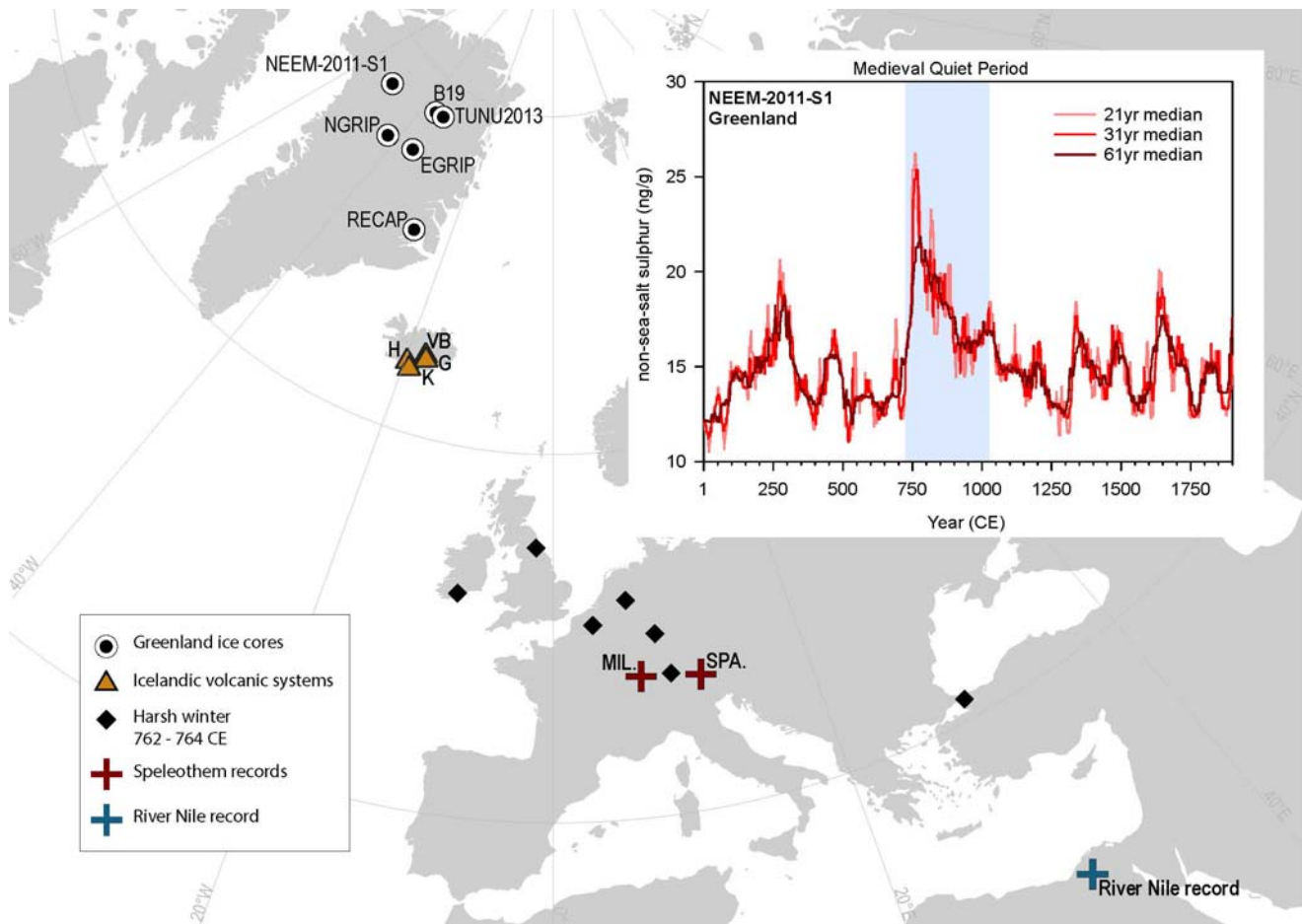
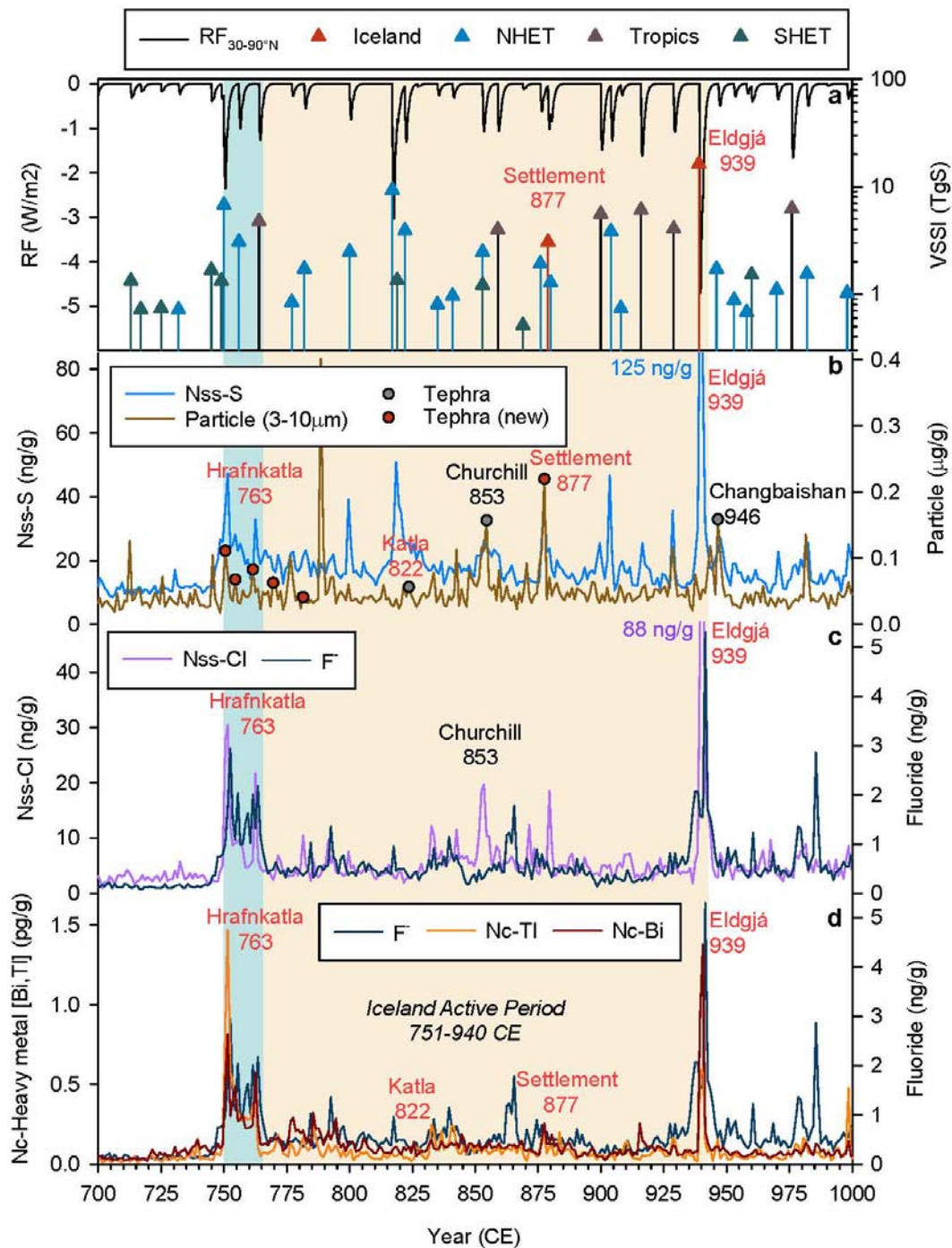
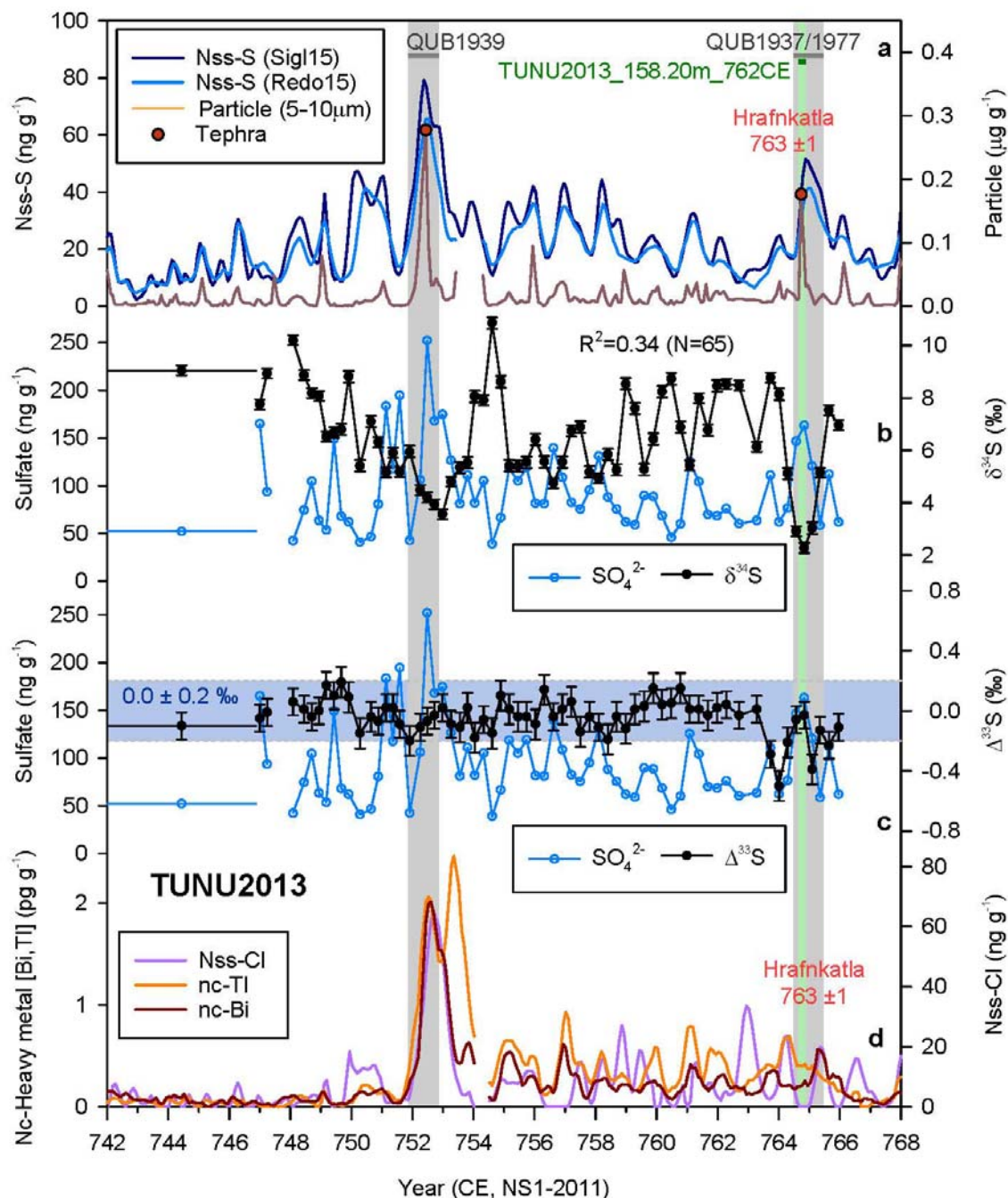


Figure 1: Cooling anomalies in the North Atlantic region: Map showing location of the Greenland ice core sites (B19, TUNU2013, NEEM-2011-S1, NGRIP, EGRIP, and RECAP) used within this study, along with the four most active Holocene Icelandic volcanic systems (orange triangles) Hekla (H), Katla (K), Grímsvötn (G), and Bárðarbunga–Veidivötn (VB). The locations of the Milandre (MIL) and Spannagel (SPA) speleothem records (red crosses) and River Nile low stand records (blue cross) utilised within this study are shown. The black diamonds denote harsh winters in the years following the 763 ± 1 CE Hrafnkatla volcanic eruption across Europe, as cited in the historical written records⁶¹. Inset figure: 21-year, 31-year, and 61-year median non-sea-salt Sulfur (nssS) concentrations for NEEM-2011-S1 across the period 1–1900 CE. Concentrations during the Medieval Quiet Period (blue shading) are unprecedented in the pre-industrial (before 1900 CE) atmosphere of the Common Era.



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809 **Figure 2: Greenland ice core volcanic tracers across 700–1000 CE:** a) Radiative forcing [30°–90°N] and VSSI (Tgs)
 810 linked to Icelandic (red triangle), northern hemisphere (NHET) (beige triangle), Tropical (black triangle), and southern
 811 hemisphere (SHET) (green triangle) volcanic eruptions; (b) Non-sea-salt Sulfur (nssS) stack (N = 6) of Greenland ice
 812 cores and associated insoluble particle record (3–10 μm) with both existing sampled tephra locations (grey) and new
 813 samples (red) shown; (c) Non-sea-salt chlorine (Nss-Cl) stack (N = 6) and Fluoride (F) stack (N = 2) of Greenland ice
 814 cores; (d) Non-crustal thallium (TI), non-crustal bismuth (Bi) stack (N = 4) and Fluoride (F) stack (N = 2) of Greenland
 815 ice cores. The blue shading denotes the Hrafnkatla episode (751–763 CE), and the beige shading shows the duration
 816 of the Icelandic Active Period (751–940 CE).



817

818 **Figure 3: Hrafnkatla episode:** (a) Non-sea-salt Sulfur (nssS) from TUNU2013 ice core and associated insoluble particle
819 record (5–10 µm) across the period 742–768 CE. Targeted tephra sample (grey) and sulfur isotope (green) locations
820 (which were screened for tephra) are shown, with the geochemically confirmed occurrences of cryptotephra
821 indicated by red circles; (b) Measured δ³⁴S (‰) and sulfur concentrations versus time recorded in the TUNU2013 ice
822 core; (c) Measured Δ³³S (‰) and sulfur concentrations versus time recorded in the TUNU2013 ice core. Blue shading
823 indicates stratospheric Δ³³S range (0.0 ± 0.2 ‰); (d) non-crustal thallium (Tl), non-crustal bismuth (Bi) and non-sea-
824 salt chlorine (nss-Cl) record for TUNU2013 ice core. A pronounced enrichment can be observed at the beginning of
825 the Hrafnkatla episode.

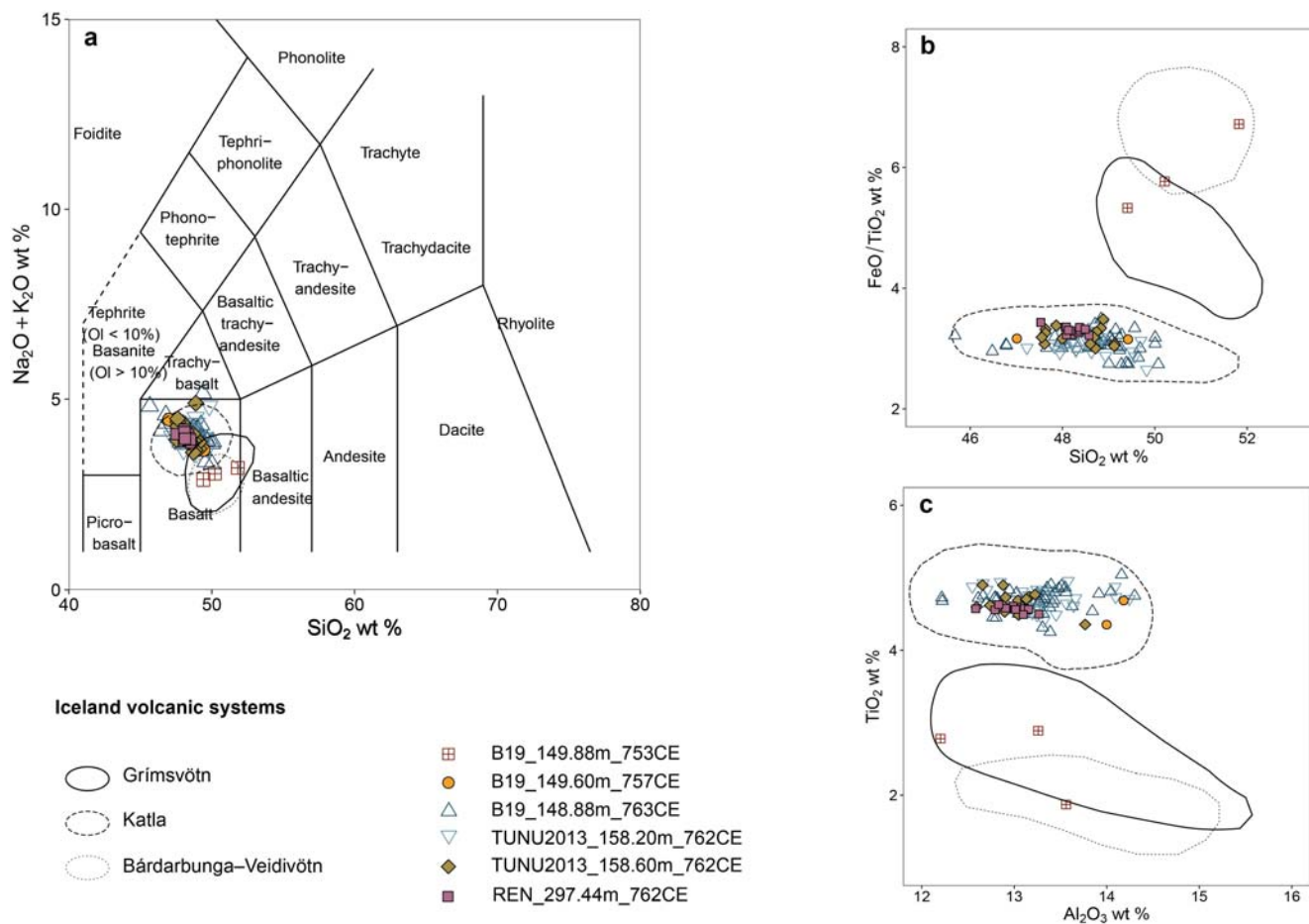
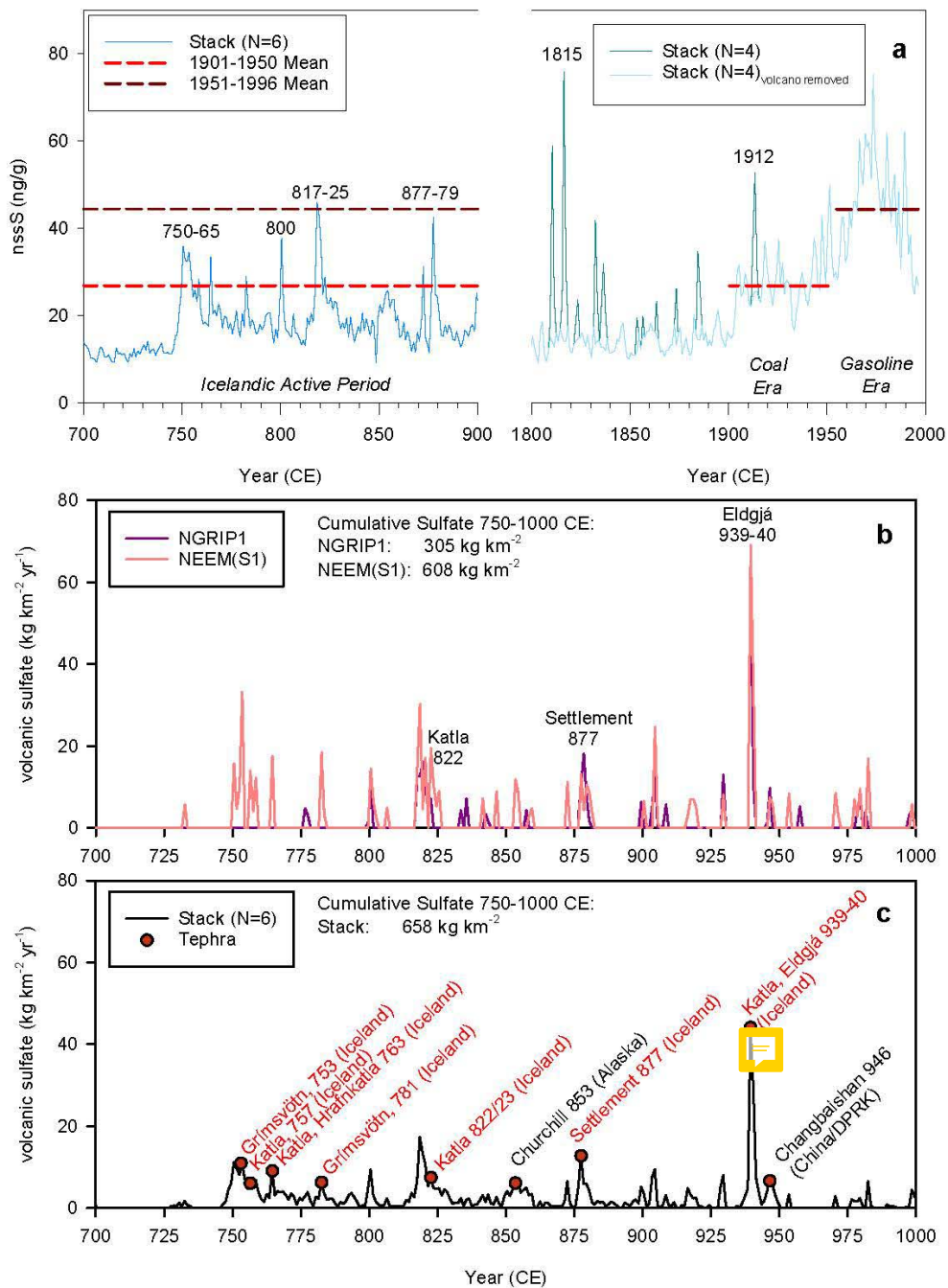
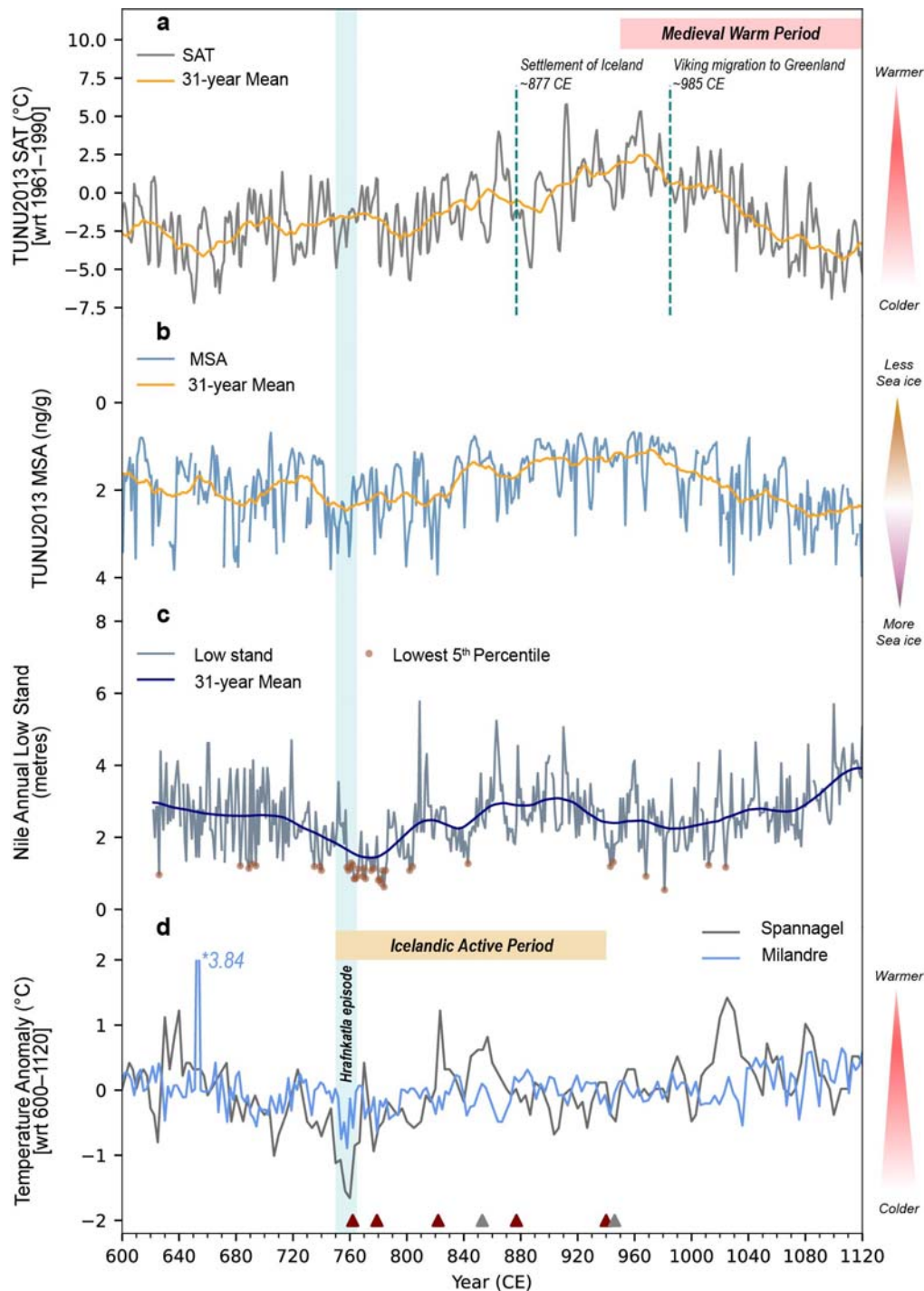


Figure 4: Pinpointing the source volcanoes of the Hrafnkatla episode: a) Major element glass shard analyses from the 751–763 CE event cryptotephra deposits from the B19, TUNU2013, and RECAP ice cores plotted based on magmatic composition- Total Alkali (Na₂O + K₂O) vs Silica (SiO₂)¹²⁵ and major element bi-plots of b) FeO/TiO₂ vs SiO₂ and c) TiO₂ vs Al₂O₃. Geochemical fields for the Grímsvötn, Katla, and Bárðarbunga-Veiðivötn Icelandic volcanic systems have been constructed based on published data^(24,126,127,128,129,130,131,132,133,134,135,136) and proximal analyses obtained in this study (see Supplementary data). All data have been normalised to 100 % (anhydrous basis).



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838 **Figure 5: Volcanic sulfate deposition in Greenland ice cores:** a) Nss-S (stack N = 6) across the period 700–900 CE and
839 1800–2000 CE (stack N = 4). Mean nss-S for the periods 1901–1950 (red dashed line) and 1951–1996 (brown dashed
840 line) are shown for comparison; b) Annual volcanic sulfate deposition from NGRIP1³⁹ (purple line) and NEEM-2011-
841 S1⁴⁰ (red line) across the period 700–1000 CE; c) Annual volcanic sulfate deposition (this study) using a stack (N = 6)
842 of Greenland ice cores, where calculated cumulative volcanic sulfate for the period 750–1000 CE is greater (658 kg
843 km⁻² compared to 305 kg km⁻² and 608 kg km⁻² for NGRIP1 and NEEM-S1 respectively). Tephra occurrences with
844 confidently attributed sources throughout this period are indicated by the red circles. A comprehensive overview can
845 be found in Coulter et al. (2012)¹³⁷.



846

847 **Figure 6: Palaeoenvironmental and proxy records 600–1120 CE:** a) TUNU2013 surface air temperature (SAT [$^{\circ}\text{C}$]
848 with 31-year mean (yellow line). The timing of the Hrafnkatla episode (blue) is shown, alongside the onset of the
849 Medieval Warm Period (red) and settlement of Iceland and Greenland (green dashed line); b) TUNU2013 MSA with
850 31-year mean (yellow line); c) Nile annual low stand with 31-year mean (dark blue line). The lowest 5th percentiles are
851 marked (brown circles); d) Spannagel cave (grey line) and Milandre cave (blue line) speleothem temperature
852 anomalies [$^{\circ}\text{C}$] with reference to mean annual air temperature across the 600–1120 CE period. Icelandic (red
853 triangles) and NHET (grey triangles) volcanic eruptions during the Icelandic Active Period (yellow) are shown.

Supplementary Information to:

“Long-lasting flare-up of volcanic activity in early medieval Iceland caused climate shocks throughout Europe”

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38 I. Supplementary Methods:

39 A) ice-core cryptotephra preparation

40 B) proximal tephra preparation

41 II. Supplementary Tables: Tables 1 - 3

42 III. Supplementary Figures: Figures 1- 11

43 IV. Supplementary References

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I. Supplementary Methods

A) *Summary of the Ice-core cryptotephra sample preparation methodology for geochemical analysis at the University of Bern adopted within this study.*

Centrifuging Samples. The ice samples were placed into individual 250ml Nalgene bottles and melted at room temperature. Samples were then gradually decanted into 15ml centrifuge tubes and centrifuged at 2500 rpm for 5 minutes to concentrate the particles at the bottom of the tube. The excess meltwater from these samples was then removed by pipette until ~1ml of water remained. These steps were repeated until all of the meltwater had been transferred from the Nalgene bottles. The bottles were then rinsed twice with ~13ml of deionised water which was then transferred to the tubes and centrifuged. This was to prevent any particles remaining in the bottle.

Mounting Samples. Acrylic discs with a 1cm diameter pre-drilled hole in the centre (total diameter 2.5cm), were covered with Kapton tape at the base and placed upon aluminium platters on top of a hotplate. The concentrated 1ml water samples were then gradually pipetted into the holes and left to evaporate at 60°C. When the samples had completely evaporated, the hotplate was switched off and the samples were left to cool for 1 hour. Buehler EpoThin™ Epoxy resin was prepared by slowly mixing hardener and resin, which was then placed into a desiccator and brought under vacuum for 3 minutes to eliminate bubbles. The resin was then slowly used to backfill the hole, covering the sample using a wooden stick. Care was taken to ensure that a dome did not form on the top of the sample. Samples were then left to cure for 72 hours at room temperature.

Identification of tephra and polishing. After the samples had cured, the Kapton tape was removed and placed on a separate labelled slide. All samples were checked for the presence of tephra using a polarising microscope at x40 magnification. Samples where tephra was identified were prepared for geochemical analysis by polishing the epoxy surface and tephra shards using a 1µm and ¼µm diamond paste for ~10 minutes. This was in order to create a flat surface for geochemical analysis.

95 B) *Summary of the proximal tephra sample preparation methodology for geochemical analysis at the University of*
96 *Bern adopted within this study.*

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98 **Sampling.** Proximal material for the candidate events was obtained from fieldwork in Iceland. Sampling locations
99 were selected based on previous published work ^{1,2,3}. Soil sections were exposed, cleaned, logged, and
100 photographed, and the candidate tephra layers were sampled and placed into labelled bags and sealed. Samples
101 were subsequently transported back to the University of Bern, Switzerland.

102 **Lab preparation.** Samples were transferred from the labelled bags into labelled centrifuge tubes. The samples were
103 then crushed until they were a fine powder. Once all samples were transferred and crushed, each centrifuge tube was
104 filled with ~14 ml of deionised water. Each sample was subsequently sieved between 125µm and 25µm, and the
105 retained residue transferred to separate centrifuge tubes for each size fraction. These were then filled with ~14 ml of
106 deionised water and centrifuged at 2500 rpm for 5 minutes to concentrate the particles at the bottom of the tube.
107 This was done in a clean environment, one sample at a time, to avoid cross-contamination. The smallest size fraction
108 (25-63 µm) was selected for analysis as it was closest in size range to the ice core tephra samples.

109 0.25ml of the concentrated sample was pipetted into the 1cm hole of the pre-drilled acrylic discs which were covered
110 with Kapton tape at the base and placed upon aluminium platters on top of a hotplate and left to evaporate at 60°C.
111 When the samples had completely evaporated, the hotplate was switched off and the samples were left to cool for 1
112 hour. Buehler EpoThin™ Epoxy resin was prepared by slowly mixing hardener and resin, which was then placed into
113 a desiccator and brought under vacuum for 3 minutes to eliminate bubbles. Resin was then used to slowly backfill the
114 hole and cover the sample using a wooden stick. Care was taken to ensure that a dome did not form on the top of
115 the sample. Samples were then left to cure for 72 hours at room temperature.

116 **Polishing.** After the samples had cured, the Kapton tape was removed and placed on a separate labelled slide.
117 Samples were prepared for geochemical analysis by polishing the epoxy surface and tephra shards using a 1µm and
118 ¼µm diamond paste for ~10 minutes. This was in order to create a flat surface for geochemical analysis.

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II. Supplementary Tables

Ice core	Longitude/Latitude	Elevation m (a.s.l)	Bottom depth (m)	Timespan [(-) BCE/CE)	Accumulation rate (m/yr)	Method (lab)	Parameters analysed	Refs
NGRIP1	75.10°N/42.32°W	2917	1351	187 to 1996	0.19	IC (PICE)	SO ₄ , Cl ⁻ , F ⁻	⁴
NGRIP2	75.10°N/42.32°W	2921	3085	[-]123000 to 1998	0.19	CFA- ICPMS (DRI)	S, Cl, Bi, TI, Cd, Part., H ⁺ , conductivity (cond)	⁵
NEEM (2011-S1)	77.45°N/51.06°W	2450	410	86 to 2006	0.22	CFA- ICPMS (DRI)	S, Cl, Bi, TI, Cd, H ⁺ ,	⁶
TUNU2013	78.04°N/33.88°W	2105	213	280 to 2013	0.1	CFA- ICPMS (DRI)	cond, tephra(UniBE)	⁷
B19	78.00°N/36.40°W	2234	150.4	748 to 1993	0.1	CFA- ICPMS (DRI)	S, Cl, Bi, TI, Cd, Br, Particles (Part.), H ⁺ , cond, tephra(UniBE)	<i>This study</i>
EGRIP	75.38°N/35.60°W	2458	Ongoing	Ongoing to 2015	0.11	IC (AWI)	SO ₄ , Cl ⁻ , F ⁻	<i>This study</i>
RECAP	71.3°N/26.7°W	2340	562.13	[-]118834 to 2013	0.45	CFA- ICPMS (DRI)	S, Cl, Bi, TI, Cd, Br, Part., tephra(PICE)	<i>This study</i>

Supplementary Table 1: Summary of ice cores utilised in this study including longitude/latitude, elevation, bottom depth, timespan, and accumulation rate. Parameters analysed which were used from each ice core within this study are shown, along with the lab where the measurements were undertaken, unless otherwise indicated. Lab abbreviations: AWI = Alfred-Wegner Institute, Germany; UniBE = University of Bern, Switzerland; DRI = Desert Research Institute, USA; Edin = University of Edinburgh, UK; PICE = Physics of Ice, Climate and Earth, Denmark; QUB = Queen's University Belfast, UK; StAiG = University of St Andrews, UK.

Event	Diagnostic Parameter	Age NS1-2011 and stated uncertainty (CE) for NEEM- 2011-S1	Age DRI_NGRIP2 and stated uncertainty (CE) for NGRIP2	Mean Age (CE)	Corresponding date from other proxy or documentary sources
Volcanic eruption 626	Sulfur increase	626 (±0)	625 (±2)	626	Documentary records of volcanic dust veil starting in October 626 ⁷
Onset Hrafnkatla Episode	Thallium increase	752 (±1)	750 (±2)	751	None
End Hrafnkatla Episode	Thallium decrease	764 (±1)	762 (±2)	763	Strong winter climate anomalies in Europe and Black Sea culminating in winter 763/764 ⁸
Solar proton event 774	¹⁰ Be increase	774 (±0)	773 (±2)	774	June to August 774, based on seasonally resolved 14C data from an interhemispheric tree-ring network ⁹
Volcanic eruption 800	Short-lived sulfur spike	800 (±1)	799 (±2)	800	Strong NH summer cooling in various tree-ring reconstructions in 800 ^{10,11}
Eldgjá eruption 939-940	Sulfur increase	939 (±0)	939 (±2)	939	Documentary records of volcanic dust veil starting in 939 ⁷ and major tree-ring cooling in 940; Nile summer flood failure in 939 ¹²

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146 **Supplementary Table 2:** Key dates in two independent ice-core chronologies from Greenland, NS1-2011⁷, and
147 DRI_NGRIP2 ⁵ the latter being identical with GICC21 between 681 and 1270 CE with GICC21¹³. Between 626 and 800
148 CE, DRI-NGRIP2 is within error identical with NS1-2011 but is consistently older by 1 to 2 years relative to NS1-2011
149 and relative to specific marker horizons identified in tree-ring and documentary records.

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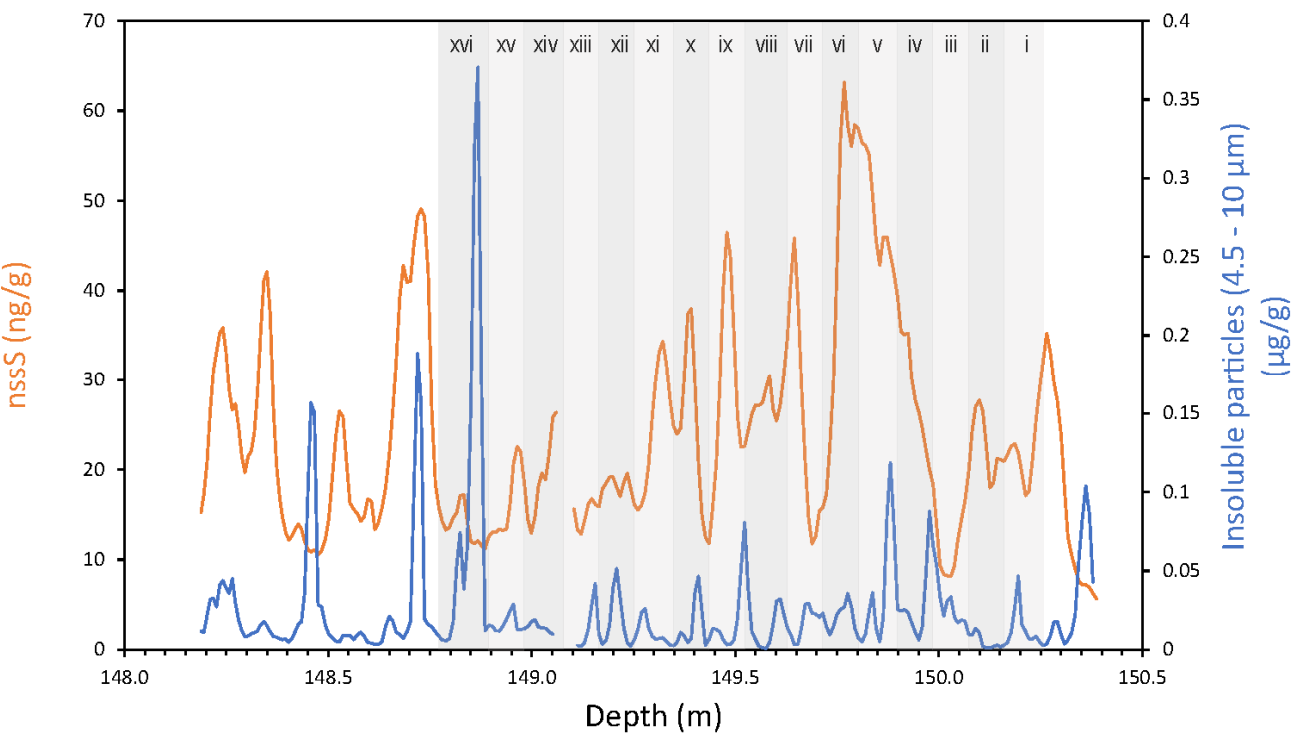
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Event	Diagnostic Parameter	Age NS1-2011 and stated uncertainty (CE) for NEEM-2011-S1	Age DRI_NGRIP2 and stated uncertainty (CE) for NGRIP2	Age GICC21 and stated uncertainty (CE) for NGRIP2	Mean Age (CE)	Corresponding date from other proxy or documentary sources
Volcanic eruption 626	Sulfur increase	626 (± 0)	625 (± 2)	626 (± 4)	626	Documentary records of volcanic dust veil starting in October 626 ⁷
Onset Hrafnkatla Episode	Thallium increase	752 (± 1)	750 (± 2)	750 (± 4)	751	None
End Hrafnkatla Episode	Thallium decrease	764 (± 1)	762 (± 2)	762 (± 4)	763	Strong winter climate anomalies in Europe and Black Sea culminating in winter 763/764 ⁸
Solar proton event 774	¹⁰ Be increase	774 (± 0)	773 (± 2)	773 (± 4)	773	June to August 774, based on seasonally resolved 14C data from an interhemispheric tree-ring network ⁹
Volcanic eruption 800	Short-lived sulfur spike	800 (± 1)	799 (± 2)	799 (± 4)	799	Strong NH summer cooling in various tree-ring reconstructions in 800 ^{10,11}
Eldgjá eruption 939-940	Sulfur increase	939 (± 0)	939 (± 2)	939 (± 4)	939	Documentary records of volcanic dust veil starting in 939 ⁷ and major tree-ring cooling in 940; Nile summer flood failure in 939 ¹²

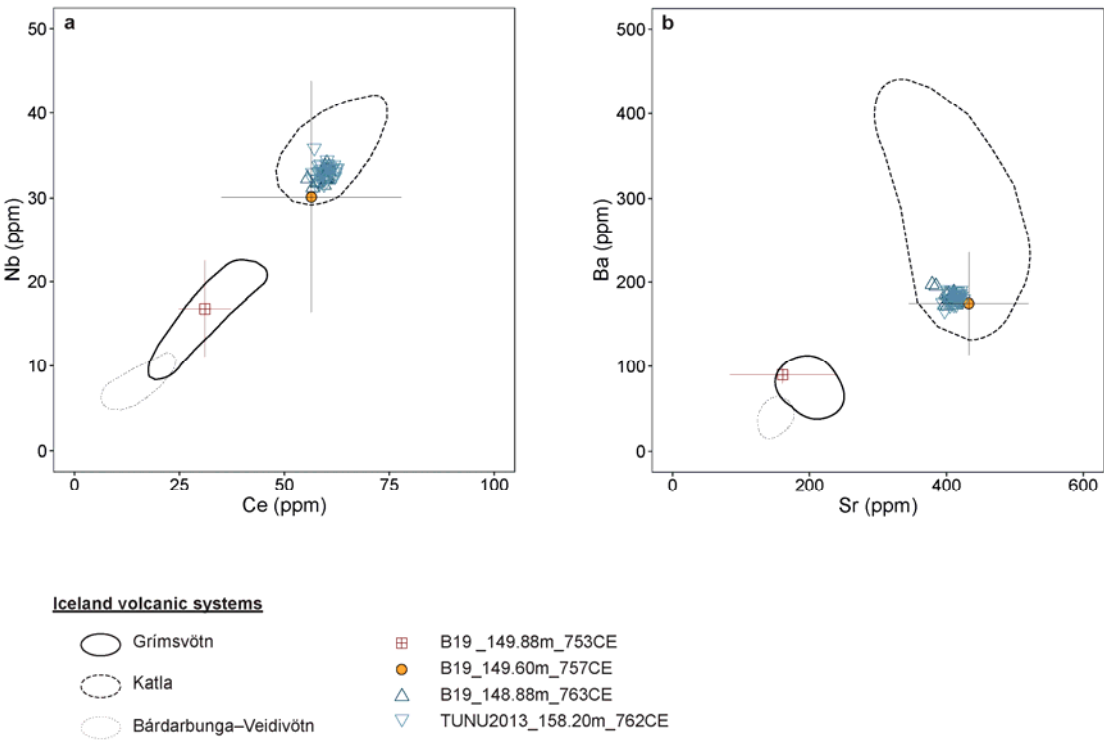
Supplementary Table 3: Key dates in three ice-core chronologies from Greenland, NS1-2011⁷, DRI_NGRIP2⁵, and GICC21¹³.



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166 **Supplementary Figure 1:** Sub-annual nssS and insoluble particle records from the B19 ice core showing the targeted
167 sampling strategy adopted. Grey bars denote depth interval of samples; (i) 150.24—150.15 m; (ii) 150.15—150.06 m;
168 (iii) 150.6—149.97 m; (iv) 149.97—149.88 m; (v) 149.88—149.79 m; (vi) 149.79—147.7 m; (vii) 149.7—149.61 m;
169 (viii) 149.61—149.51 m; (ix) 149.51—149.42 m; (x) 149.42—149.33 m; (xi) 149.33—149.24 m; (xii) 149.24—149.15
170 m; (xiii) 149.15—149.06 m; (xiv) 149.06—148.97 m; (xv) 148.97—148.88 m; (xvi) 148.88—148.76 m.

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185 **Supplementary Figure 2:** Trace element bi-plots a) Nb vs Ce and b) Ba vs Sr for 751—763 CE event cryptotephra
186 deposits from B19 and TUNU2013 ice cores, based on absolute concentrations (ppm) from single grains. Geochemical
187 fields for the Grímsvötn, Katla, and Bárðarbunga–Veidivötn Icelandic volcanic systems have been constructed based
188 on published data (^{14,15,16,17,18,19,20}) and proximal analyses obtained in this study (see Supplementary data). Error bars
189 represent 2 σ standard deviation of individual analyses in relation to the ML3B reference glass.

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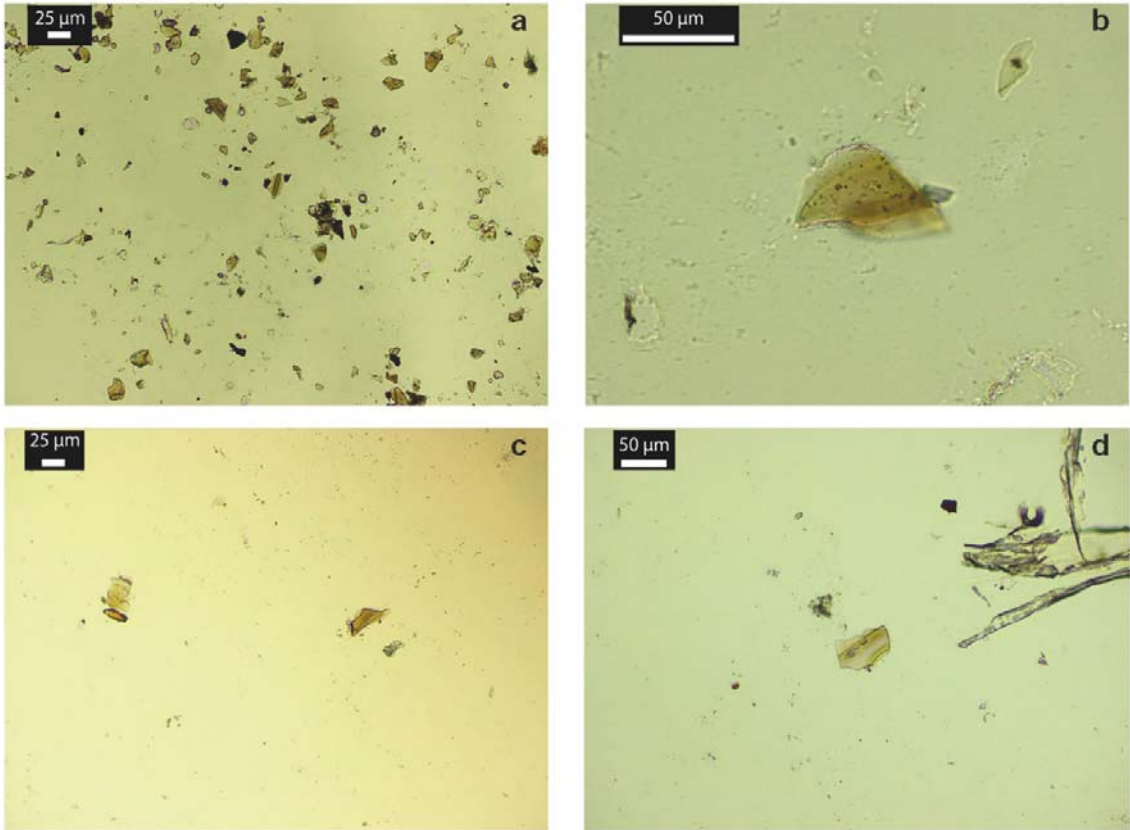
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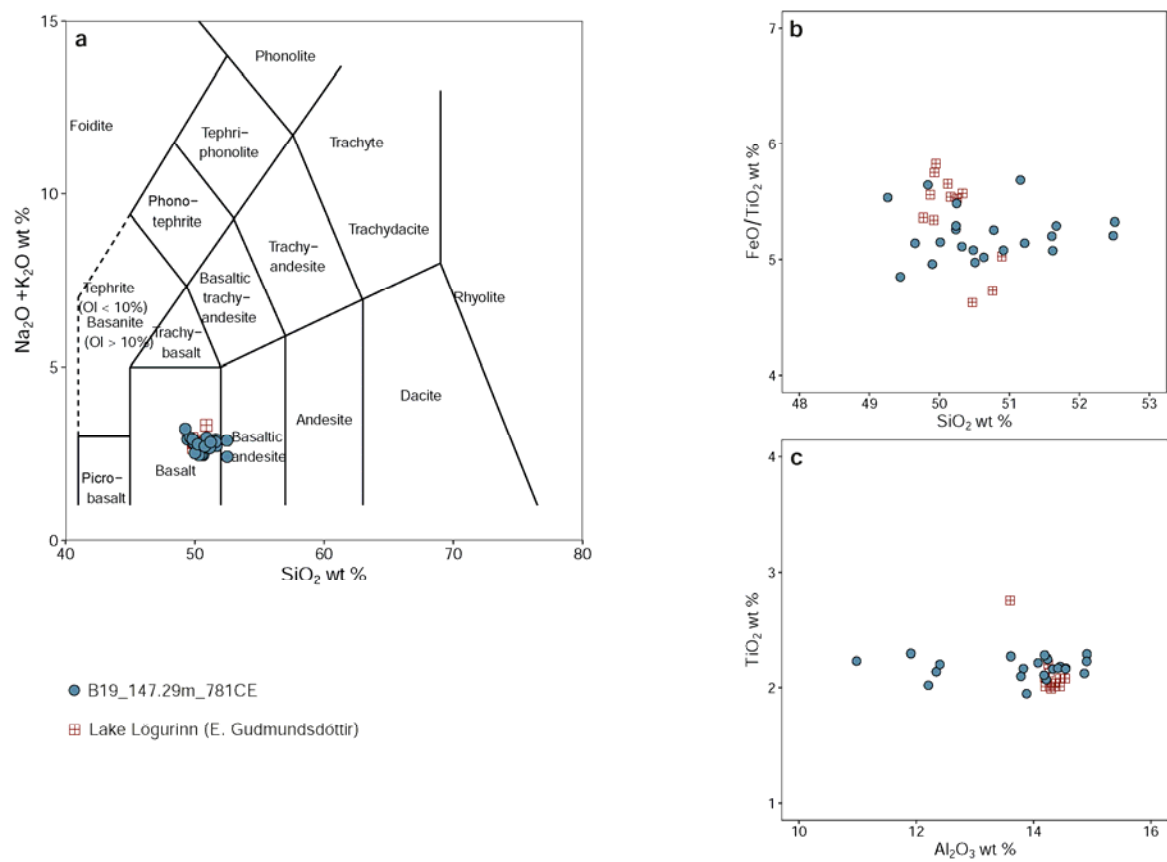
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Supplementary Figure 3: Photographs of glass shards identified in B19_148.88m_763CE (a, b) and TUNU2013_158.2m_762CE (c, d) ice core samples which have been geochemically correlated to the Hrafnkatla volcanic eruption (Katla, Iceland).



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221 **Supplementary Figure 4:** a) Major element glass shard analyses from the 781 ± 2 CE event cryptotephra deposits
222 from the B19 ice core plotted based on magmatic composition- Total Alkali (Na₂O + K₂O) vs Silica (SiO₂)²¹ alongside
223 existing analyses from an associated horizon in lake Lögurinn, Iceland (Pers. Comm. E. Gudmundsdóttir). Major
224 element bi-plots b) FeO/TiO₂ vs SiO₂ and c) TiO₂ vs Al₂O₃. All data have been normalised to 100 % (anhydrous basis).

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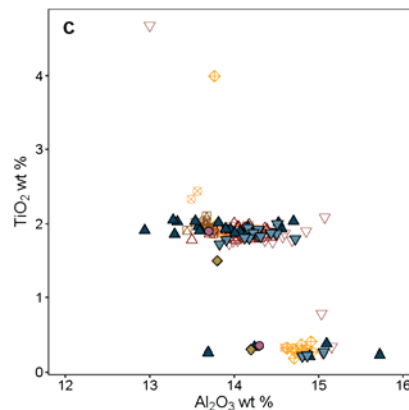
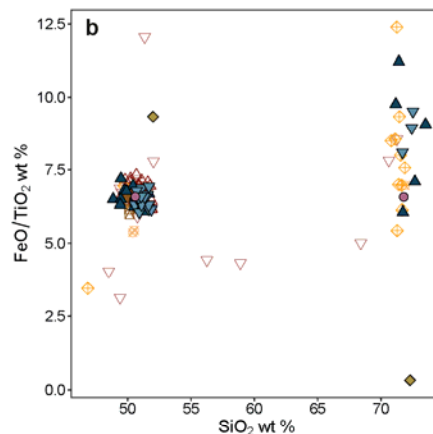
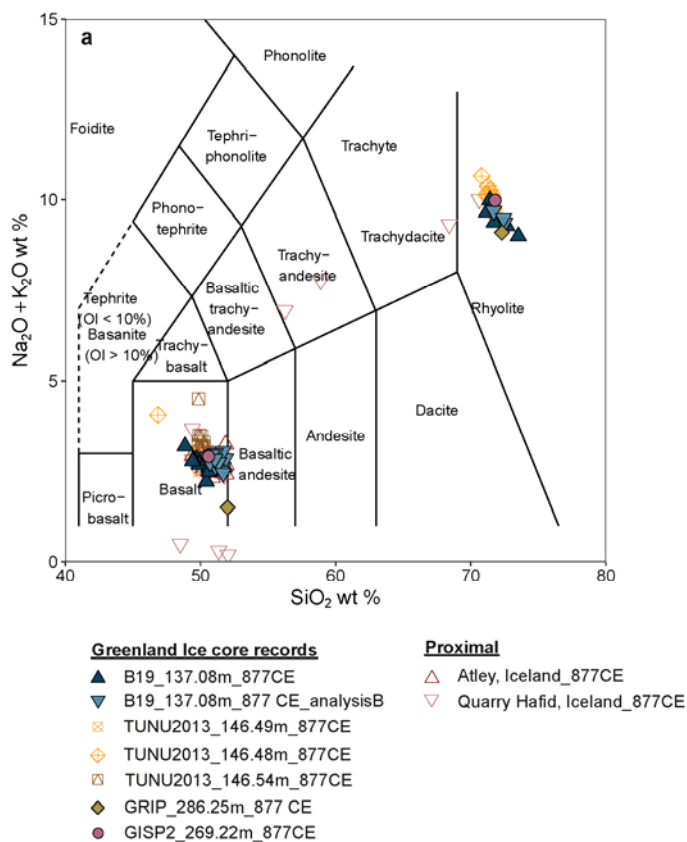
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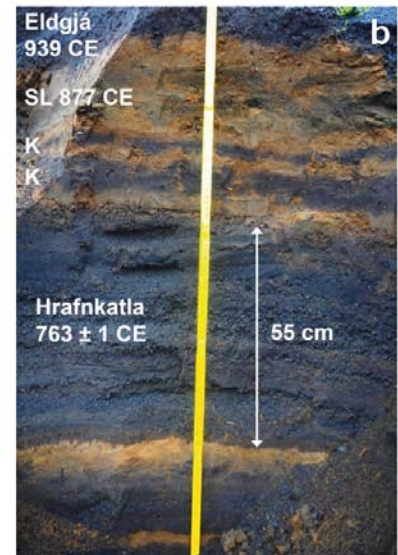
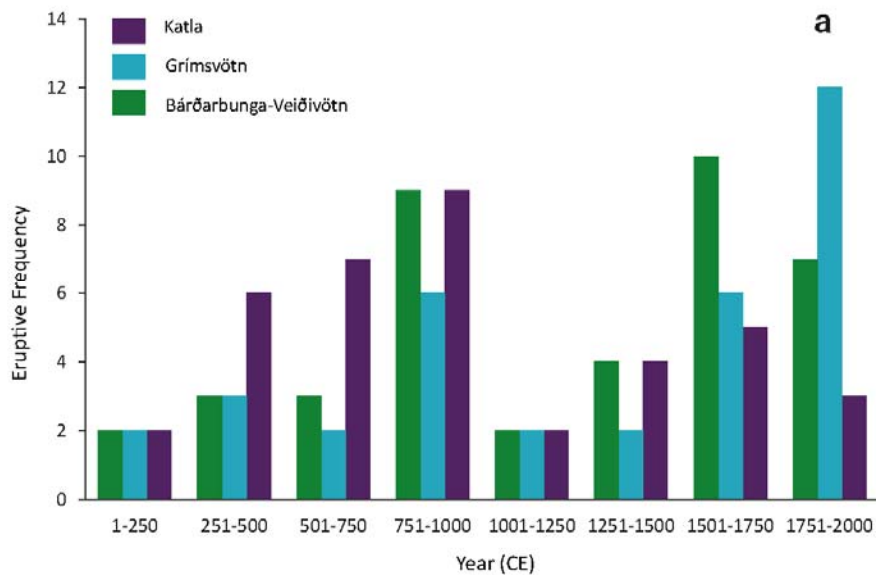
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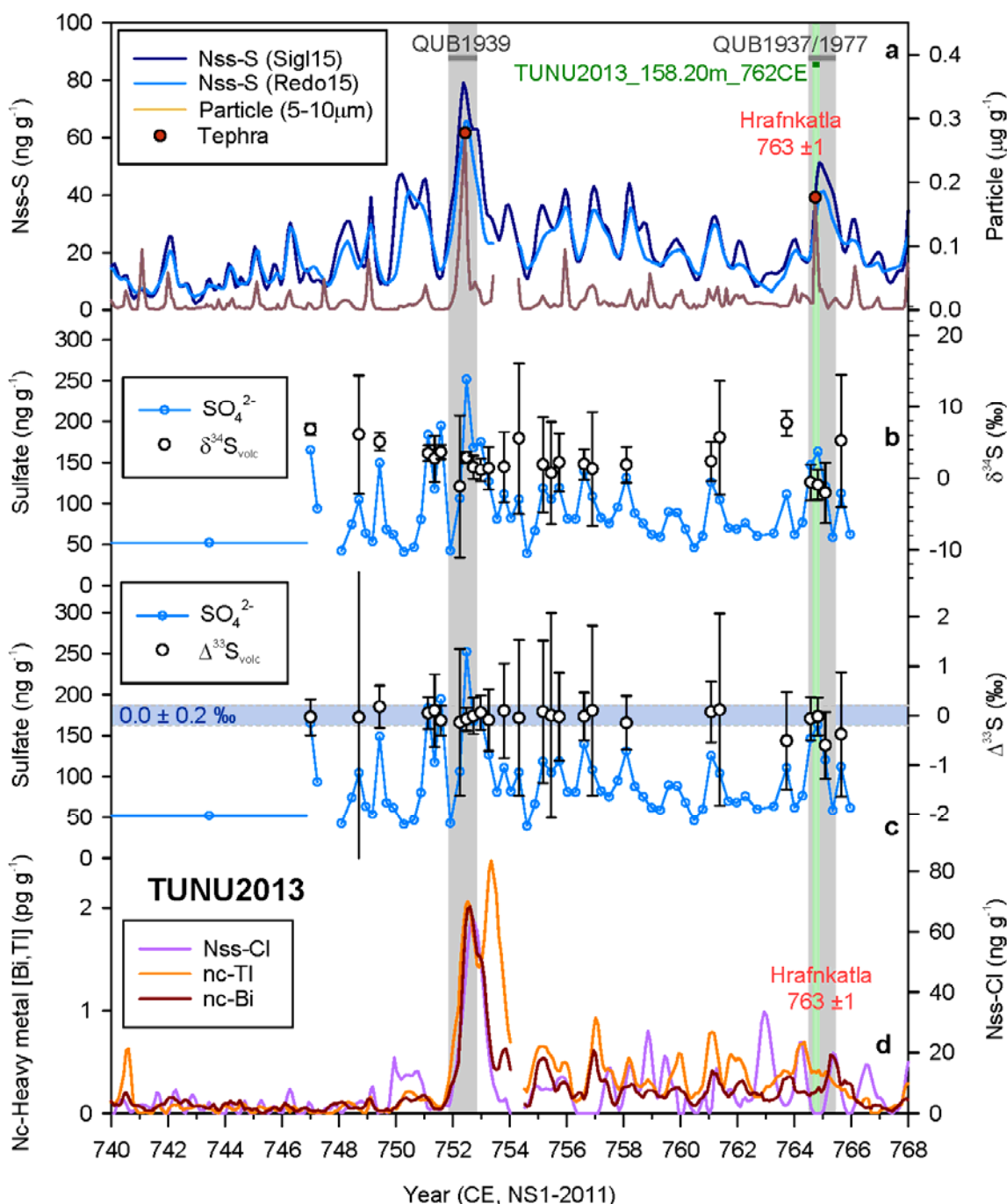
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Supplementary Figure 5: New major element glass shard analyses from the 877 CE event cryptotephra deposits from the B19 and TUNU2013 ice cores plotted based on magmatic composition – Total Alkali ($\text{Na}_2\text{O} + \text{K}_2\text{O}$) vs Silica (SiO_2)²¹ alongside published ice core analyses (GRIP²², GISP2²³, TUNU2013²⁴) and proximal material from the same event collected as part of this study (see Supplementary data). Major element bi-plots are shown in b) FeO/TiO_2 vs SiO_2 and c) TiO_2 vs Al_2O_3 . All data have been normalised to 100 % (anhydrous basis).

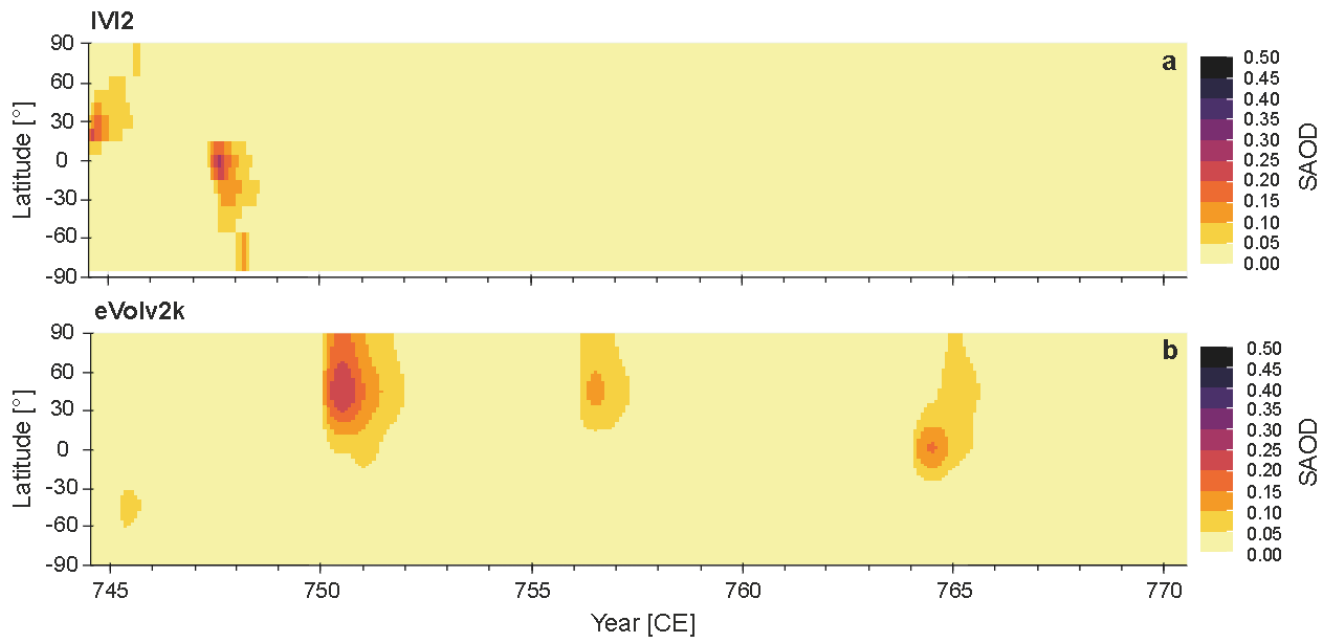


Supplementary Figure 6: a) Plot showing estimated eruptive frequency of Katla, Grímsvötn and Bárðarbunga-Veiðivötn volcanic systems during the last 2000 years in 250-year time windows, determined using the Catalogue of Icelandic Volcanoes²⁵. An increase in eruptive frequency from all volcanic systems is observed during the period 751–1000 CE; b) Annotated photograph showing proximal soil profile near Atley, Iceland [63.67295°N, 18.68231°W] encompassing the Icelandic Active Period. Correlated tephra horizons are marked, with the Hrafnkatla event (763 ± 1 CE) defining the base of profile and the Eldgjá eruption (939 CE) defining the top.

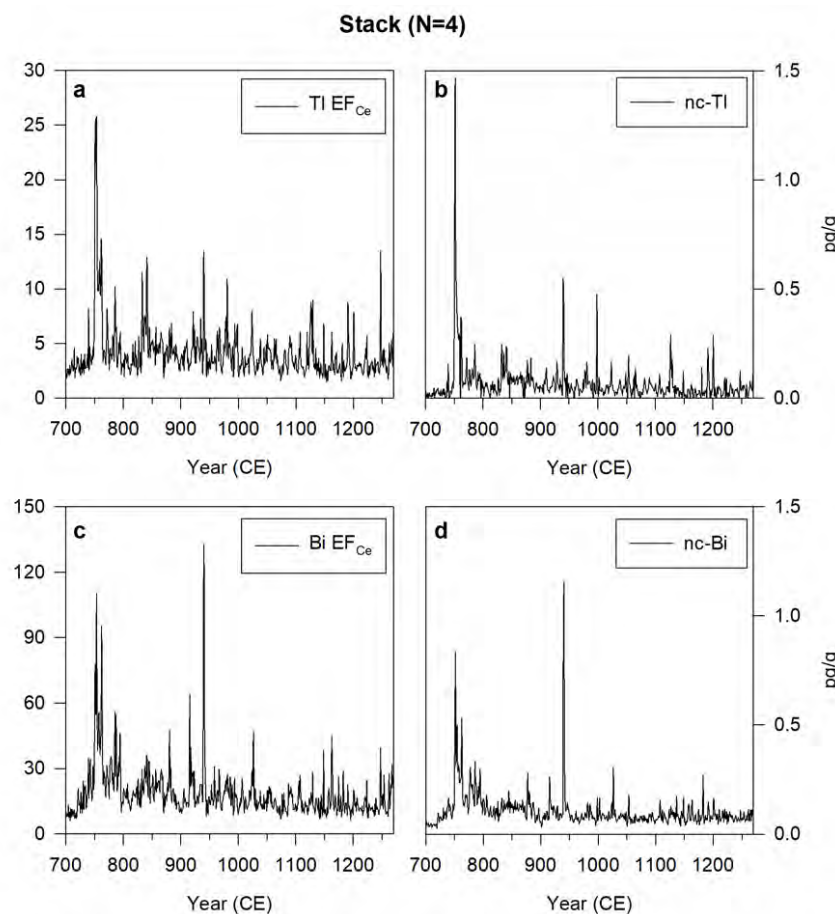


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264 **Supplementary Figure 7:** (a) Non-sea-salt Sulfur (nssS) from TUNU2013 ice core and associated insoluble particle
 265 record (5–10 μm) across the period 742–768 CE. Targeted tephra sample (grey) and sulfur isotope (green) locations
 266 (which were screened for tephra) are shown, with the geochemically confirmed occurrences of cryptotephra
 267 indicated by red circles; (b) Background corrected $\delta^{34}\text{S}$ (‰) and sulfur concentrations versus time recorded in the
 268 TUNU2013 ice core; (c) Background corrected $\Delta^{33}\text{S}$ (‰) and sulfur concentrations versus time recorded in the
 269 TUNU2013 ice core. Blue shading indicates stratospheric $\Delta^{33}\text{S}$ range (0.0 ± 0.2 ‰); (d) non-crystal thallium (Tl), non-
 270 crystal bismuth (Bi) and non-sea-salt chlorine (nss-Cl) record for TUNU2013 ice core. A pronounced enrichment can
 271 be observed at the beginning of the Hrafnkatla episode.



Supplementary Figure 8: a) IVI2²⁶ and b) eVolv2k²⁷ Stratospheric Aerosol Optical Depth (SAOD) estimates based on polar ice core records across the period 745—770 CE. No volcanic events were detected in the IVI2 estimates across the Hrafnkatla episode, in contrast to eVolv2k where some volcanic perturbations were incorporated. Data was plotted using the SAOD Visualization App (<https://cj.argpages.usask.ca/saod-visualization/>) Ogilvie, C.J. [2022].



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295 **Supplementary Figure 9:** a) Enrichment Factor of Tl relative to Ce (a crustal dust tracer) defined as EF
296 $= ([\text{Tl}]_{\text{ice}} / [\text{Ce}]_{\text{ice}}) / ([\text{Tl}]_{\text{crust}} / [\text{Ce}]_{\text{crust}})$ with crustal values from McLennan (2001)²⁸; b) non-crustal-Tl defined as $[\text{Tl}] -$
297 $(0.01172 \times [\text{Ce}])$; c) Enrichment Factor of Bi relative to Ce calculated as for Tl; non-crustal-Bi defined as $[\text{Bi}] -$
298 $(0.00198 \times [\text{Ce}])$. All Cerium concentration values were corrected for under-recovery during CFA analysis by multiplying with
299 1.7 following Arienzo et al.²⁹.

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IV. Supplementary References

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Reviewer #1 (Remarks to the Author):

Dear editor and authors

Thank you for the opportunity to review this work. In summary, the authors present a very rich dataset, and make interesting and novel observations and discoveries about volcanism in Iceland in the period 700-1000 CE and the associated climate impacts. The scope of the study makes it suitable for publication in this journal. There are many things that the authors do well, but there are also numerous issues, and I recommend revisions before publication. I don't believe additional analysis is necessary but the manuscript would need substantial revision in a few places.

Focus, content, and interpretation:

(1)- I found that the title, the abstract, and the introduction are somewhat misrepresentative of the main focus and content of the study. They claim that the focus of the study is on the so-called Icelandic Active Period (IAP, 751-940 CE), during which there were multiple eruptions. The authors do indeed present some data about several eruptions; however, the lion's share of the results and discussion, and the data-rich figures are on one eruption only, Hrafnkatla, which occurred at the beginning of IAP. Why did the other eruptions within this period did not receive the same level of detailed analysis and discussion as Hrafnkatla? There are other very significant eruptions during this time period, including the Settlement event, and of course, the very large Eldgjá eruption, which is at least as large as Hrafnkatla and maybe even larger. It therefore seems strange (or at least, unexplained) to focus on Hrafnkatla only, especially since the title, abstract and introduction promised to focus on the other eruptions too. I'm sure the authors have reasons for choosing to focus on the one eruption (perhaps the data set for the other eruptions was not as detailed?) but in this case, the title, abstract and introduction should not be overpromising and should be rewritten accordingly. The results about this one eruption are still very interesting and warrant publication.

Thank you for your comment. A focus was made on the Hrafnkatla eruption as this marked the beginning of this abrupt onset in sulfate observed within the Greenland ice cores. The other larger events during this time period (Settlement Layer 877 CE and the 939 CE Eldgjá eruption) have previously been identified within the Greenland ice core records and received considerable attention (Plunkett et al., 2023 ¹; Oppenheimer et al., 2018 ²). The scope of this study was to focus on the detection of prolonged volcanic emissions (i.e., the Hrafnkatla episode) within the ice-core records, thereby reconciling existing mismatches between the proximal Icelandic geological records and global volcanic aerosol radiative forcing estimates. Furthermore, documentary evidence and paleoenvironmental archives from across Europe cite the winters following the Hrafnkatla event (at the end of these prolonged emissions) as being particularly severe. We therefore wanted to explore a possible volcanic cause for such cooling and considered the Hrafnkatla eruption as a likely mechanism. However, we emphasise that this link would need to be further explored via modelling, which is beyond the scope of this study. Following your suggestion, we have rewritten the title, abstract, and introduction to more accurately represent the focus and content of this study.

(2) - One of the most interesting results of the paper is that the new results disagree with previous work about the prevalence of Icelandic volcanism during the studied time period. But this is not explored or explained in the Discussion. Please include a discussion of this and give some possible reasons for the disagreement.

A more detailed discussion for the causes of this disagreement has now been included in the discussion.

(3) - Figure 5: I didn't understand why the peak values are lower on panel c) compared to b) since the stack is reported to be higher overall in c). Please explain the reasons for this in more detail in the main text. This seems to be a potentially interesting observation?

The differences in the peak values between panel B (single ice cores) and panel C (stack of 6 ice cores) can be explained by a number of factors, the two most important ones are 1) the relative age uncertainty within the stack of the six ice cores; 2) the relative small annual snow accumulation rates (e.g. for B19, TUNU2013, EGRIP) and lower measurement resolution (e.g. EGRIP) of some of the additional ice cores used in the stack composite. Both factors result in a peak broadening (e.g. from post-depositional effects like snow-mixing) and thus will lead to reduced amplitude of the peaks in the stack, in comparison to single ice cores at high snow accumulation sites such as NEEM or NGRIP. In addition, site elevation and distance between the ice-core site relative to major volcanic centres result in different sensitivities to volcanic sulfur emissions. We have provided information about small remaining age uncertainties in the ice cores (LINES: 67-71; 356-366 and their different sensitivities to source emissions in the manuscript. (LINES: 185-188)

Manuscript structure and organisation:

The manuscript is very data-rich and could do with being structured in a better way to help the reader to navigate it.

(4) - Manuscript would benefit greatly from a introduction section on each of the target eruptions. Including but not limited to: size and eruption style; magma composition; which volcanic system they occurred in; tephra size distribution; any information about plume height; length of eruption; etc. Some of this information is included later on, in the Results, but it is required earlier on to be able to better understand the text, and especially the figures.

Thank you for the suggestion. A table (Table 1) has been added in the introductory section providing information regarding all events that are discussed in Figure 3. Eruptions which are discussed in greater detail within this study are indicated by the asterisk.

(5) - There is a large number of supplementary sections (tables, text, figures) and these are not referenced properly in the main text, making it very hard to find the relevant one. Typically the text just says “data is in supplement” with no reference to specific section. Please remedy this.

Thanks, all supplementary data has now been labelled appropriately.

(6) - Most figures have several panels. Some of the panels are mentioned and discussed in the text; others are not, or only mentioned in passing. Please make sure that all panels are discussed adequately, otherwise it is not clear what they are contributing to the discussion.

Thanks, we have adjusted this and removed panels which were not discussed adequately within the text. Former Figure 2 panel (a) has been removed and part of this data (radiative forcing) is now shown as the new Figure 1 panel (a).

Methods and referencing:

(7)- The methods are detailed for most sections (see minor comments on the manuscript file). The section on proximal tephra sampling is, however, missing critical information such as sampling locations, eruption layers which were sampled, references for locations of each, etc.

Thanks, we have now provided more detail regarding the proximal tephra sampling methods, with an additional Table added in the supplementary Tables (Supplementary Table 4), a map of Iceland showing the sampling locations (Supplementary Figure 10), and annotated photographs of the outcrops (Supplementary Figure 11).

(8) - Data availability was not satisfactory. In the Methods, it says “all raw data are available in the Supplement” but there was no raw data in the supplement. There were figures, but these are not raw data. Data have to be made available in a spreadsheet or another format where other people can analyse or plot it up. Similarly, it says in the Acknowledgements that raw data will be uploaded to

a repository but this had not been done. I appreciate that the authors may want to embargo the raw data but it should be available to the reviewers during the review process.

Apologies, there was difficulty in uploading the supplementary data file to the tracking server. The data has been generated in many different laboratories. All the new data has now been uploaded by the data generators to either PANGAEA (<https://www.pangaea.de/>) or the Arctic Data Center of the National Science Foundation's Office of Polar Programs (<https://arcticdata.io/>). We will provide for each data publication the corresponding DOI upon receipt in the Data availability section, next to all other published data used in this study. We are aware that processing times of the data submissions can be up to a few months and have therefore added the relevant data to the supplementary data file to support the reviewers assessments. We are willing to provide any data needed by the reviewers for their assessments.

(9) - Referencing is poor in places, in particular within figure captions. Some of the figures present data from other studies but references are not given.

Thanks, this has now been adjusted.

Figures:

- Generally good but see some comments on the manuscript file

Best wishes,

Dr Evgenia Ilyinskaya

University of Leeds

Specific comments (copied by author's from the commented PDF file)

Main text:

(10) Line 21-22: unclear whether you mean that individual eruptions produce SO₂ for a few years, or if that's the atmospheric lifetime of SO₂. Needs clarifying

The abstract has been rewritten to better reflect the focus of the main text. This sentence has now been removed – see reply to comment 1.

(11) Line 35: eruptions, not volcanoes

The introduction has been rewritten to better reflect the focus of the main text. This sentence has now been removed – see reply comment 1.

(12) Line 41-42: why are anthropogenic sources not listed under different sources?

See reply to comment 11.

(13) Line 42: awkward phrasing because you are saying "sulfur from volcanic eruptions may also be sourced from volcanic eruptions"

See reply to comment 11.

(14) Line 58-60: The disagreement with previous work is one of the most interesting results of your paper but it is not really discussed or explained. Please include this in your discussion and give some possible reasons for the disagreement

Thanks, see reply to comment 2

(15) Line 69: Figure is missing latitude values

Thanks, the figure has been redrawn (new Figure 2), with latitude values made more visible.

(16) Line 71: clarify that the figure shows non-sea salt sulfur

The inset figure 1 has now been moved to form part of the new figure 1 as panel b). The figure caption has been corrected to say that it shows non-sea-salt sulfur.

(17) Line 105: in which section? there is a lot of information in the supplementary material and it is difficult to know which section the authors are referring to

See reply to comment 8.

(18) Line 113-118: This information should be included in the introduction, or soon after, because it helps the readers understand the references to different eruptions on the figures and in the main text

A Table (Table 1) has been added to the introductory section – see reply to comment 4.

(19) Line 150: Awkward phrasing. If background contains 0.0 volcanic sulfate it is awkward to say that "background consists of volcanic sulfate"

Thanks. Our statistical outlier detection methods to estimate volcanic sulfate contributions cannot resolve minor but frequent volcanic sulfur contributions from smaller volcanic eruptions (i.e., VEI 3 and smaller). Therefore, time-periods with ice core indicated depositions of 0 kg km^{-2} of volcanic sulfate may still include volcanic contributions from such small eruptions, as suggested by Jongebloed et al. (2023)³ which is based on sulfur isotope analyses. We have added a sentence in the methods section (Volcano detection) to clarify that background sulfate still contains some sulfate of volcanic origin.

(20) Line 157: reference needed

Reference added.

(21) Line 198: When did this event occur? give dates, even if it is not known with certainty

Event date of ~17.7ka added following McConnell et al. (2017) ⁴ and Mason et al. (2022) ⁵.

(22) Line 205: It would be even more appropriate to refer to findings of Ilyinskaya et al 2021 that looks at differences in dispersion patterns of major and trace elements

<https://www.nature.com/articles/s43247-021-00146-2>

Thank you, we have added this reference according to your suggestion.

(23) Line 215: the more commonly accepted term is flood basalts

Thank you, this has been changed.

(24) Line 238-241: Pinatubo was a VEI 6, was Hrafnkatla also a VEI 6? If not, then the difference in eruption strength and plume injection height are also a key control on the differences in climatic patterns

This sentence has been rewritten to provide better comparison.

(25) Line 352: please refer to specific sections of the supplementary material to make it easier for the reader to locate the relevant information. This comment applies throughout the manuscript

See reply to comment 5.

(26) Line 369: I cannot see any raw data in the Supplement

See reply to comment 8.

(27) Line 385: I do not see any trace metal data in the .docx file that was made available to me.

See reply to comment 8. Trace metal data has now been included the Supplementary Data file.

(28) Line 435: flux of which species? clarify and elaborate on whether there are any uncertainties associated with assuming the species. For example, if you are calculating SO₂ flux from sulfate, you are assuming a conversion rate

This has been amended in the text. We have now specified that we mean the time integrated sulfate mass deposition at the ice core site.

(29) Line 483-485: All data should be available to reviewers during the review process

See reply to comment 8.

(30) Line 809-811: give references for data in figure A in this caption since it is not your original data

This panel (a) has now been moved to Figure 1 panel (a) with references added.

(31) Supplementary information:

Line 95-96: because of the high number of different analysis done in this study, it would be very helpful for the reader if you would list in this section which analyses were done on these samples

We have added an additional Analysis section in the Supplementary Methods (B) to clarify this.

(32) Line 98: this section needs more detail where exactly the samples were collected, and from which eruptions exactly. A table would be useful

Thanks, done. Supplementary Table 4 and annotated photographs of soil sections (Supplementary Figure 11) and map of sampling locations (Supplementary Figure 10) have been added. Further details of the Atley site can be found in Óladóttir et al. 2005⁶, with this reference additionally added to the list of published work in the sampling sub-section (Supplementary Methods B).

Reviewer #2 (Remarks to the Author):

The paper focusses on a 150-yr period (751–940 CE) during which there are relatively high levels of non-sea-salt sulphur in Greenland ice cores. Data are presented for various non-sea-salt sulphur peaks to suggest a higher frequency of volcanic activity from Iceland during this time. The paper then attributes the odd climate anomalies experienced in Europe/North Africa in the time period to the long-lived Icelandic Hrafnkatla eruption (751–763 CE).

(33) It is clear from existing Icelandic records that there was more volcanic activity from Iceland between 750 and 940 CE. The new data from the ice cores on this activity is only part of the record, with other studies providing additional key evidence. The new aspect of this study is the link between the volcanism and impact, and this is very speculative. The title of the paper is not justified as the only impact discussed is related to the Hrafnkatla eruption. Tying the Hrafnkatla episode, with stratospheric sulphur injection only at the end (~763 CE), to the cooling recorded in the European speleothem (Fig. 6d) is tenuous. The cooling recorded commences before 760 CE and so cannot be attributed to the stratospheric injection associated with eruption. In addition, the link to the Nile low stands is also weak as there are other periods when it is low. These linked impacts need to be more critically reviewed and require more discussion.

Thanks for your comment. The title of the paper has been amended – see Reviewer 1, comment 1, reducing the emphasis on the climate impact in the title. The introduction has been rewritten to better frame the focus of this study, which was to investigate this period of increased background sulfate within the Greenland ice-core records, thereby resolving mismatches between proximal Icelandic geological records and global volcanic radiative forcing estimates. We focus on the possible climatic impacts of the Hrafnkatla episode, which marks the beginning of the Icelandic Active Period, as this is where the abrupt increase in sulfate occurs in the Greenland ice-core records. We explore the possible climatic impacts of prolonged volcanic emissions, such as the Hrafnkatla episode, as documentary evidence of severe winters coinciding with Hrafnkatla episode are recorded across

Europe. The exceptional cooling of circa 10 years in both speleothem records occurs prior to the onset of the stratospheric end-phase of this episode. Owing to the resolution of the speleothem records (~ 1 sample for every two years) and uncertainty across this time-period (~ 16 years) it is difficult to align to a precise date. However, due to the date obtained from historical records, which agree with the ice-core records, we consider the cooling anomalies observed in two central European speleothem records to be a result of the Hrafnkatla eruption.

Although there are other periods where the Nile low stand is low, we wanted to highlight the clustering of low stands during and following the Hrafnkatla episode. We suggest that this clustering maybe due to feedbacks associated with the prolonged volcanic emissions attributed to Iceland, with the Nile having a demonstrated sensitivity towards Icelandic volcanic eruptions such as Eldgjá 939 CE or Laki 1783 CE (Oman et al., 2006) ⁷. We have further added a few key publications (Booth et al. 2012 ⁸; Chiang et al. 2012 ⁹; Hwang et al. 2013 ¹⁰; Allen et al. 2015 ¹¹; Brönnimann et al. 2015 ¹²) from a large body of literature focusing on the sensitivity of the ITCZ and tropical precipitation to volcanic and anthropogenic sulfate emissions during the 20th century (LINES: 277-282). Additionally, we have now emphasised that there are great uncertainties in feedbacks associated with prolonged volcanic emissions and that these may be better explored in future work (i.e., climate model studies).

(34) The paper relies heavily on information presented in both the supplementary information and the supplementary data. The new data, such as the glass chemistry and evidence for the provenance of the tephra needs to be in figures and data tables in the paper. Furthermore, the figure that shows the frequency of eruptions from Iceland, based on the Icelandic database, is key to the main conclusion and should also be in the paper. The paper needs reorganisation to accomodate the data plots and tables.

Thanks for the suggestion. The new glass data has been added to Figure 4, with individual plots for each event found in the Supplementary Information (Supplementary Figures 2, 5, and 6). All geochemical data can be found in the Supplementary Data, and is now fully referenced within the

main text, as per the Editor's suggestion. The eruption frequency plot (formerly Supplementary Figure 6) has now been moved to the main text as Figure 1c.

Comments related to specific sections of the paper and SI:

(35) Line 99 – see comment below on Supplementary Fig. 3.

Done.

(36) Line 102 – state the volume of the sample processed.

We have provided the cross-sectional area rather than volume, as this is a better representation of primary ash fall due to the nature of the ice cores. Samples with a greater volume do not necessarily equate to more tephra. However, if you increase your cross-sectional area, there is a greater chance that tephra from an event may be identified.

(37) Line 115 – typically referred to as bulk volume.

Thanks, this has been corrected.

(38) Line 188 – this implies that the episode could be related to activity from other systems in addition to Kafla. Evidence needs to be presented or cited if this is the case.

We assume that you are referring to line 118? We provide evidence of activity of other Icelandic volcanic systems within this study through the geochemical attribution/correlation to the Grímsvötn volcanic system in 753 CE.

(39) Line 122 – what is the evidence to attribute this to secondary deposition? This requires more detail and discussion.

As the 766 ± 1 CE sample (B19_148.67m_766CE) contained a number of other particles (i.e., minerals) in addition to the analysed tephra shards. The other particles observed in the sample could

have only come to the ice sheet via secondary transport processes, and as the tephra shards are of a similar size, this suggests that they many have been transported to the site via similar processes. To further test this, additional geochemical analyses (including trace element) may help to better constrain this. We have now clarified this within the main text.

(40) Line 129: This figure and the data should be in the paper.

Major geochemical data regarding the Settlement Layer is now presented in Figure 4, alongside all other major analyses obtained in this study. A separate, detailed plot regarding the Settlement Layer data can be found in Supplementary Figure 6.

(41) Line 133: Reference should be inserted.

Thanks, reference added.

(42) Line 137: The new information presented is not enough evidence to indicate that Iceland is highly active and productive in the period. It needs a lot more context before such a conclusion is reached. The next sentences provide the evidence and are the foundation for this comment so should be moved earlier. Supplementary Fig 6 should be referenced and moved into the paper.

This paragraph has been rewritten/restructured to provide more context as per your suggestion (LINES: 133-142). Supplementary Figure 6 has now been moved to the main text forming part of Figure 1 as panel (c).

(43) Fig. 1. Would be good to have an inset over Iceland showing all the volcanoes that are known to be active during this period and where samples were taken.

We thank the reviewer for this suggestion. A map showing sampling locations and volcanoes with known activity during the period 700-1000 CE has now been added to the supplementary information (Supplementary Figure 10).

(44) Fig. 2. Hard to distinguish SHET and NHET on (a) as the colours are too similar, and they are not the same as mentioned in the caption, e.g. tropics are grey not black. References for the tephra samples in (b) that are not new (in grey) should be added to the caption. Not sure why F- is in both (c) and (d). The caption should have 'Nc-Tl' and 'Nc-Bi' in brackets instead to be consistent with the figure.

Thanks. We have redrawn Figure 2 (now Figure 3 in main text) removing panel (a). References for the old tephra samples have been added in the caption. F⁻ has been removed from the heavy metal panel (panel c). The caption has been updated to be consistent with the figure, as per the reviewers suggestion.

(45) Fig. 3. caption for (d): abbreviations in brackets are not the same as those used in the legend on the figure.

Thanks, we have adjusted this.

(46) Fig. 4. Plot the compositions of the other tephra layers identified, e.g. all the new layers shown in Fig. 2b, and fields for the different Icelandic volcanoes.

As per the reviewer's suggestion, Figure 4 now presents the major geochemical data for all events obtained within this study, with Icelandic volcano geochemical fields shown. Separate plots for each event can be found in the Supplementary Information.

Comments on the Supplementary Information:

(47) Line 69, cryptotephra samples: This is missing the detail of what samples were selected, how they were selected, and what length of time a sample represents. It is mentioned elsewhere but it should also be stated here.

We have added a subsampling section to Supplementary Methods (A) to clarify this.

(48) Line 98, Sampling: I think this information needs to be in the main paper. It would be good to have annotated photographs or logs. The sampling points should be plotted on a map.

Thanks for the suggestion, but we have left the proximal tephra sampling methods in the Supplementary Method section, as this was not the main focus of the study. An annotated photograph of each sampled outcrop can be found in Supplementary Figure 11 and sampling locations are shown in Supplementary Figure 10, as per your recommendation – see reply to comment 43.

(49) Line 166-167: Sampling strategy for what measurements? In line 99 of the manuscript, it implies that samples were taken “...by exactly co-registered analyses of size resolved insoluble particle concentrations and sulphur” but on this figure (which is referred to) it seems that sampling was continuous. This needs to be better explained as suggested above (comment for line 69). It would be helpful if there was an additional x-axis with dates so it can easily be related to other figures.

This figure previously showed the sampling approach adopted for the Hrafnkatla episode where the overlap of background samples either side of the main peak conveyed a continuous approach, rather than a targeted approach. We have selected a different event (877 CE Settlement Layer) to more clearly show how sampling was undertaken to the reader. Dates have also been added, as per the reviewers suggestion.

(50) Line 185, S.Fig.2: It is not clear why only fields for those volcanic systems are shown. This needs further justification. I assume because the Iceland record is already well known, e.g., Gudmundsdottir et al., (2016). Other volcanic systems, such as Snaefellsjökull, Askja and Torfajökull, could be shown for comparison. Why are the volcanic fields not marked on S.Fig.4 and S.Fig.5?

Only the fields for the Grímsvötn, Katla and Bárðarbunga-Veiðivötn systems are shown on the trace element plots as major element geochemical analysis allowed us to narrow it down to the Grímsvötn

and Bárðarbunga-Veiðivötn systems. The absence of volcanic fields on the additional major geochemical plots is an oversight on our part and has now been amended.

(51) Line 204, S.Fig.3: It would be great if images of the other cryptotephra shards could be added.
Done.

(52) Line 254, S.Fig.6: This is critical to the argument that Iceland is more active during the highlighted period and thus should go in the main paper.
Done – now Figure 1c – see reply to comment 41.

(53) Line 254, S.Fig.7: This appears the same as Fig. 3 in the paper. It is not clear why it is here.
Supplementary Figure 7c) and d) show the background corrected $\delta^{34}\text{S}$ and $\Delta^{33}\text{S}$, as stated in the figure caption, whilst Figure 3c) and d) show the raw $\delta^{34}\text{S}$ and $\Delta^{33}\text{S}$. This supplementary figure is included for the readers comparison.

(54) Line 277, S.Fig.8: Would be good to mark the position of the event on the figure.
Done – we have marked on the duration of the Hrafnkatla episode.

(55) Line 295, S.Fig.9: Need to mark on positions of the eruptions.
Done - We have marked on the position of the Hrafnkatla episode and Icelandic Active Period with shading. Icelandic eruptions are indicated by red triangles.

Supplementary data comments:

(56) Tab 1: This should be a table in the paper. Need to define abbreviations used, e.g., McC18; UniBe (IG). The shard concentrations for T197 and T199 need to be listed if they are Icelandic as it is directly relevant to this study and not be acceptable to just cite an in prep. publication.

Thanks for the recommendation. Following the editor's suggestion, we have left this as a table in the supplementary data, but this table is now fully referenced within the main text. Abbreviations have been defined. Shard concentrations have been listed for T197 and T199.

(57) Tab 2: This information needs to go as a table in the paper. Define fa-mI, and fa-ca. Need to add photographs or logs of the sections.

Thanks for the recommendation. Following the editor's suggestion, we have left this as table in the supplementary data, but this is now fully referenced within the main text. Abbreviations have been defined. Annotated photographs are available in the supplementary information – Supplementary Figure 11.

(58) Tab 4. Operations conditions: For what? Probably better to include in the paper or SI.

These are the operating conditions for the electron microprobe at the University of Bern. This has been clarified in the Readme tab.

(59) Tab 5. A summary table of the major element compositions should go as a table in the paper. Following the editor's suggestion, we have left this as table in the Supplementary Data, but this is now fully referenced within the main text.

(60) Tab 6. A summary table of the trace element compositions should go as a table in the paper.

See reply to comment 58.

(61) Tab 9. Define MATT. This dataset needs explanation.

This dataset has been fully defined in the Readme Tab of the data file.

Reviewer #3 (Remarks to the Author):

(62) The authors presented a new and interesting idea: An increase in Icelandic volcanism influenced climate conditions in Europe around 700-1000 CE. I think it is a very complex and multidisciplinary paper that combines paleoclimate research, tephrochronology, and glaciochemistry. Probably because of this complexity I found this paper relatively hard to read and follow. Certain parts of the manuscript probably can be reorganized. For example many details from the methods sections could be organized as tables with data and maybe most of the text can be moved to the Supplementary section. I found that certain parts of the research can not be replicated. The authors mentioned R scripts used for statistical correlations of tephra and described statistical methods for background and volcanic peak selections, but they are not provided in the paper.

Thank you for your comment. No statistical analyses were performed for the tephra geochemical correlations – R, the statistical software, was just selected to visually plot the data. This sentence has now been amended to avoid confusion. The statistical methods for background and volcanic peak selection are well established. In the methods section, “Volcano detection”, we provide an overview of these methods, and highlight alterations to thresholds and running medians used within this study. We fully cite the methods on which these are adapted from in the methods section (Traufetter et al., 2004¹³; Ferris et al., 2011¹⁴; Sigl et al., 2013¹⁵).

(63) I think that the information on methods for tephra sample extraction, preparation and measurements is well organized and reported. Probably tephra related plots would benefit with more information about why Icelandic sources are selected. For example, the possible range of major and trace element values for Iceland and other regions in Europe, Kamchatka, Alaska or Japan could be discussed in figure captions.

Thank you for your comment. We mention in the text how Icelandic eruptions have dominated the Greenland ice-core records throughout time, largely due to their proximity, which is why when initially processing the data, we focused on Iceland. However, as shown in figure 3b, other Northern

hemisphere high-latitude sources have contributed to the Greenland ice core records throughout this period (Plunkett and Pilcher 2018¹⁶; Plunkett et al., 2023¹). Within this study however, as correlations throughout the study period have been made with Icelandic volcanic systems, we have only included these geochemical fields.

(64) When I made a plot using (Sigl et al., 2022) data set, I noticed an increase in measured S values in the NEEM-2011-S1 ice core, around the 700-1000 CE interval. I also identified an offset in S levels around 820 C.E. and several increases, for example, around 1450 C.E. or 1700 C.E.. Because there is no information included on measured uncertainties, it is not clear if these changes are related to measurements or part of the environmental signal. It is specifically required by this journal to access and track all uncertainties. I suggest including uncertainties and also new plots for chemical species (e.g., S, heavy metals and halogens) affiliated with volcanic sources that would show that the 700-1000 CE interval variability is different from other intervals in the last 2000 years. Another potential problem is related to possible changes in chemical species sources or intensities of air mass transport. Such changes are sometimes driven by climate fluctuations impacting atmospheric circulation patterns. In the past, these questions were answered by applying Principal Component Analysis (PCA) to large glaciochemical data sets. A similar test could help to capture changes in volcanic species associated with S and reject possible changes in atmospheric dust transport from the Sahara region that for example has a lot of Fe, Ca and S.

Sulfur concentrations in Greenland ice cores can vary for different reasons in the pre-industrial period; 1) Clustering of large volcanic eruptions, as for example around 1450 C.E. or 1700 C.E; 2) long-term increases in volcanic activity, as discussed for the first time in this manuscript, e.g. 700-1000 CE. These changes represent an environmental signal since they are common to different ice cores (e.g., N = 5) measured with different analytical techniques (see Figure 1 below). Measurements of sulfate and sulfur in particular, are highly reproducible with typical quantification errors in the order of ~20% or better. See Supplementary Figure 7 panel (a), showing two independent analyses of

nss-S on adjacent ice sections analysed in 2013 and 2015. Analytical details on analysis techniques and replication including species such as sulfur, chlorine, bismuth, and thallium can be found in McConnell et al. 2002¹⁷; McConnell and Edwards 2008¹⁹; McConnell et al. 2017⁴ (in particular, Figures S1 to S3 in the SOM - <https://www.pnas.org/doi/10.1073/pnas.1705595114#supplementary-materials>) and McConnell et al. 2019¹⁸. Regarding atmospheric circulation changes, our approach is rather insensitive to changes in atmospheric circulation due to the large network of ice cores used. Furthermore, we do not find any significant long-term changes in typical sea-salt and dust tracers between 700-1000 CE to be indicative of changes in atmospheric circulation. We have performed Principal Component Analysis (PCA) and found three major principle components; PC1 is dominated (43% variance explained) by a source with high-loading in sulfur and heavy metals; PC2 is dominated (21% variance explained) by a source with high-loading in sulfur and no heavy metals; and PC3 is dominated (11% variance explained) by a source with high-loadings in mineral dust and typical sea-salt species (e.g., Na, Ca) (see Figure 2 below). Overall, these findings support our interpretation in that between 700 and 1000 CE volcanic sulfur emissions rich in heavy metals peaked (as shown in PC1), whereas PC2 shows single large eruptions, and PC3 does not show any long-term trends. PCA was performed on annual averaged and monthly averaged data and log transformed data. While fully consistent with our findings, we do not feel that this PCA significantly strengthens our findings.

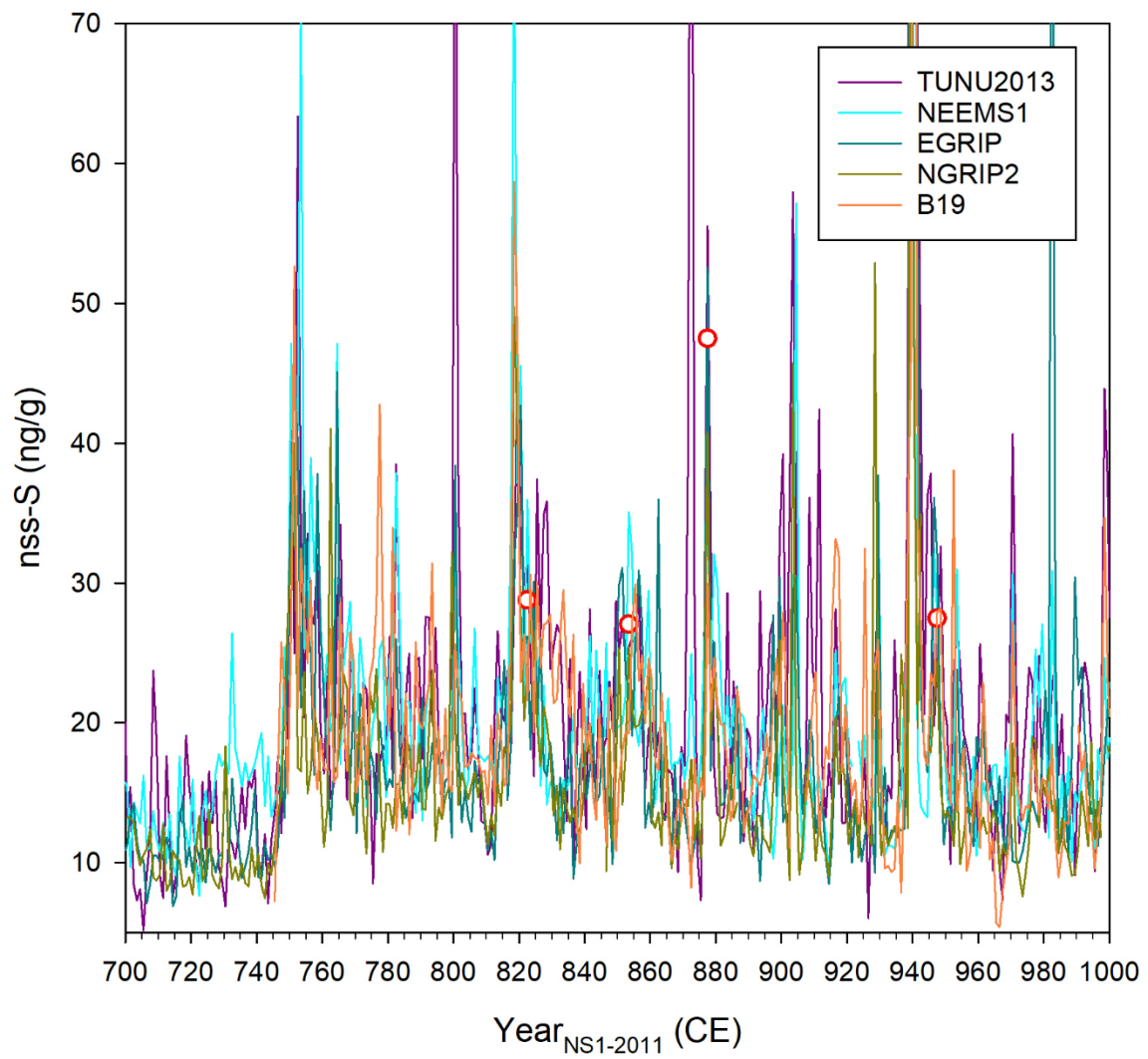


Figure 1: nss-S record from five Greenland ice-core records (TUNU2013, NEEM-2011-S1, EGRIP, NGRIP2, and B19) measured at two different labs using two different analytical techniques. The reproducibility of the signal across the five ice cores can be observed.

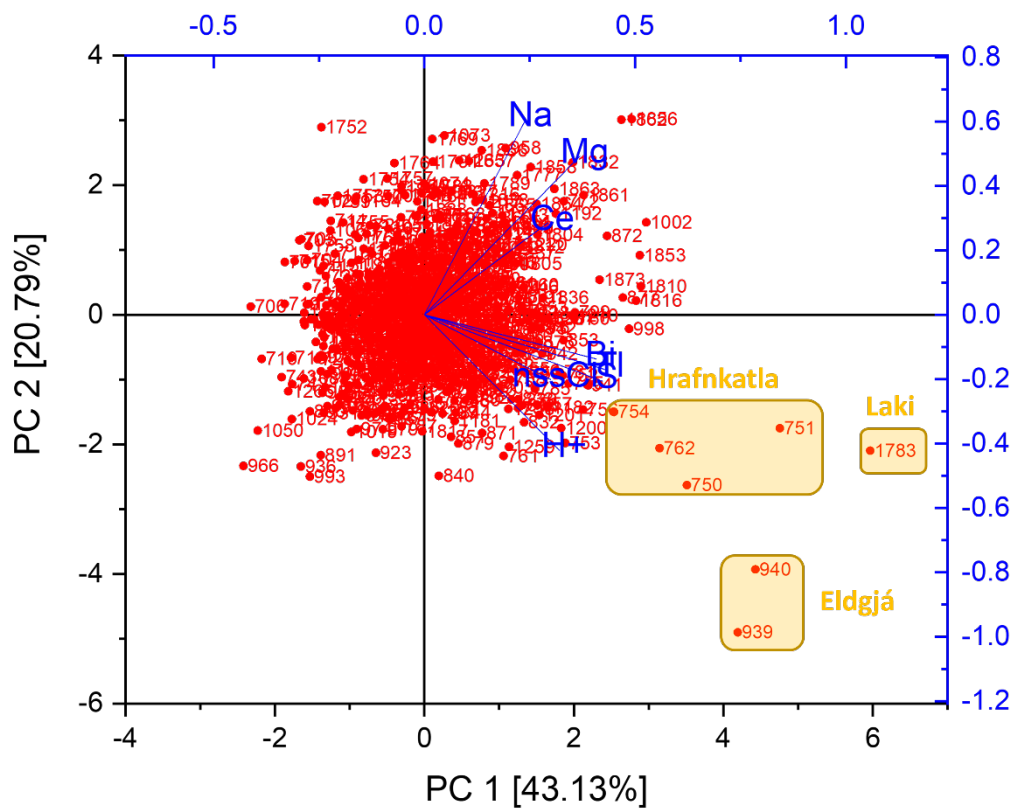


Figure 2: Bi-plot showing the scores of all 696 years for PC1 and PC2 (red) and the loading for the eight elements for PC1 and PC2 (blue). Ice-core concentration Stack (N = 4); NGRIP2, NEEM-2011-S1, B19, TUNU2013 (log-transformed); Na, Mg, Ce, Bi, Ti, S, nss-Cl, and H⁺ (annual mean) 696 years total (700-1269, 1750-1875 CE). 3 components (75% of variance); PC1 (43%: S, Ti, Bi all >0.4 loading), PC2 (21%: Na, Mg all > 0.49 loading), PC3 (11%: S, H⁺ all >0.44 loading).

(65) Finally I am not fully convinced that several unusual climate extremes in Europe could qualify as “climate shocks throughout Europe” in the title. History of global volcanism is not well developed yet and we could be missing information on some major explosive volcanic eruptions that are not associated with Iceland. These tropical events, as was demonstrated by the manuscript co-authors in the past, could be responsible for climate extremes during this interval.

The title has been amended – see reply to Reviewer 1, comment 1, reducing the emphasis on the climate impact in the title. The history of global volcanism is well-developed and based on numerous

ice core records from both hemispheres. The focus of this study was on a period where no major tropical volcanic eruptions have been identified within the ice-core records. In particular, the period 700-1000 CE is characterised by the absence of major tropical eruptions. Therefore, the stratospheric explosive Icelandic eruption of Hrafnkatla dated to 763 ± 1 CE is most likely for the trigger for the cold winters historically documented throughout Europe between 762 and 764 CE. This is consistent with other extreme winters documented following Icelandic volcanic eruptions in 822 CE, 939 CE and 1783 CE (McCormick et al., 2007²⁰; Oppenheimer et al., 2018²; Zambri et al., 2019²¹). Further analysis (i.e., climate modelling) would be helpful to explore these links.

(66) In summary, I think this interdisciplinary paper topic is suitable for the journal. I also think the idea to find evidence of a long term increase in Icelandic volcanism around 700-1000 CE in Greenland ice cores deserves proper consideration.

At its present form most of the conclusions are not fully supported by rigorous data analysis that can be replicated by the reader. I would recommend that ice core data will be presented and evaluated for a longer time frame (e.g. the last 2000 years) and it has to be demonstrated statistically, with reported uncertainties, that the interval around 700-1000 CE has unusual patterns in glaciochemistry that exceed variability for major chemical species that are associated with volcanic sources.

Thank you for the suggestion. Ice core records of sulfur (NEEM-2011-S1, TUNU2013) and sulfate (NGRIP) are publicly available (Plummer et al., 2012²²; Sigl et al., 2015²³; Sigl and McConnell 2022²⁴ - see data availability section for details) allowing for independent verification that the period 700-1000 CE is unique in the context of the last 2000 years as shown in Figure 1b. Bismuth, Thallium, Sulfur and chlorine records have been published for many ice core sites in Greenland between 1750 and 2000 CE (e.g., McConnell and Edwards, 2008¹⁹; Chellman et al. 2017²⁵; Zhai et al. 2021²⁶), which encompasses the Laki 1783 CE eruption in Iceland. Most of the data can be found in the Arctic data Centre repository. The ice core data during other time periods is not the subject of this study.

I am convinced that the data analysis I suggested is possible and it would reinforce the proposed connections between Icelandic volcanism and increases in some chemical species in Greenland ice cores. I think this alone will start a new chapter in future research that could identify and possibly quantify complex connections between volcanism and climate.

Specific Comments:

Abstract

(67) Volcanic eruptions are generally considered short-lived events, with atmospheric SO₂ emissions limited to a few years.

Comment. Volcanologically it is not a correct statement. Maybe mention that short lived emissions are common for large explosive eruptions.

The abstract has been rewritten to better reflect the focus of the main text. This sentence has now been removed – see reply to Reviewer 1, comment 1.

(68) It would be useful for the authors to define the term “climatic”. Single episodes observed across Europe and north Africa were short extreme weather events and probably will not qualify as changes averaged over 30 years of weather (the authors can define different criteria to help separate climate from weather).

The abstract has been rewritten to better reflect the focus of the main text – see reply to Reviewer 1, comment 1. We have now defined the term climatic within this study in the discussion.

Introduction

(69) It is not just sulfate aerosols (or sulfuric acid aerosols) that are influencing the state of the climate system after volcanic eruptions. It is also tephra and water. It would be useful to define specific types of emissions from volcanoes that the authors are trying to connect with the proposed increase in chemical concentrations measured in Greenland ice cores. They are using Pinatubo as an

example but it is confusing because it was a large explosive caldera forming eruption with a relatively short duration (3-5 months of climatic episode).

The Introduction has been rewritten to better reflect the focus on the main text. This sentence has now been removed – see reply to reviewer 1, comment 1.

(70) It is not clear why Holocene was selected as a target.

“To gain insights into volcanic eruptions and their associated climate impacts over longer time periods (i.e., the last 11,500 years).

The period 11,500 years was selected as an example over which past volcanism can be reconstructed. This has now been amended to e.g. rather than i.e..

(71) I would recommend that the authors include this reference to a seminal work on sulfur species in ice cores: Legrand, Michel R. 1997. “Ice-Core Records of Atmospheric Sulphur.” Philosophical Transactions of the Royal Society B: Biological Sciences 352 (1350): 241–50.
<https://doi.org/10.1098/rstb.1997.0019>.

Thanks for the suggestion. The reference was included as reference 10 in the previously submitted manuscript (reference 21 in the revised manuscript).

(72) This statement: “Most commonly, non-volcanic background concentrations” is not correct. Only after biogenic and marine sources of sulfate are removed spline or median smoothing is used to evaluate background levels of sulfate.

Thanks for your comment. This has now been amended.

(73) The references 7 and 12 are not very relevant:

“However, geological records show that this assumption may not hold true, with evidence of eruptions lasting for several decades to centuries 7,12”

A better reference is:

Self, Stephen, Millard F Coffin, Michael R Rampino, and John A Wolff. 2015. "Chapter 24 - Large Igneous Provinces and Flood Basalt Volcanism." In *The Encyclopedia of Volcanoes (Second Edition)*, edited by Haraldur Sigurdsson, 441–55. Amsterdam: Academic Press.

<http://www.sciencedirect.com/science/article/pii/B9780123859389000249>.

Thanks for the suggestion. The reference has been added.

Data sources section

(74) I think the authors did a remarkable job of coalescing different data sources. It would be helpful to the reader if existing tables clearly show ice cores, measured chemical species, and current locations of data. I spent a lot of time trying to find data and understand how the data was synthesized.

Thanks for your comment. The DOI for datasets used have now been provided (in review) - see the data availability section. See reply to reviewer 1, comment 8. We also copied these datasets into the Supplementary Data file.

References

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19th Feb 24

Dear Ms Gabriel,

Your manuscript titled "Multi-decadal changes of volcanic aerosol emissions from early Medieval Iceland detected using ice-core tracers" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Dear authors

Well done on a thorough revision of this manuscript. I am happy to recommend it for publication. I only have one minor comment on Figure 1c: “eruption frequency” axis title is missing number of years. Is the “frequency” defined as number of eruptions/year? Number of eruptions/100 years?

Best wishes,
Evgenia Ilyinskaya
University of Leeds

Reviewer #2 (Remarks to the Author):

I looked over the rebuttal and the changes to the manuscript in relation to my comments. I am happy with how they have addressed and rebutted my comments.

Reviewer #3 (Remarks to the Author):

The authors have done a thorough job revising the paper based on my previous comments. Their changes and new additions have improved the organization and data presentation in the manuscript. I recommend publishing this paper.

Dear Reviewers,

Thank you for your recommendation for publication and constructive feedback throughout the review process.

Kind Regards,

Imogen Gabriel (on behalf of all co-authors)

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Dear authors

Well done on a thorough revision of this manuscript. I am happy to recommend it for publication. I only have one minor comment on Figure 1c: "eruption frequency" axis title is missing number of years. Is the "frequency" defined as number of eruptions/year? Number of eruptions/100 years?

Best wishes,
Evgenia Ilyinskaya
University of Leeds

Thank you for your comment. We have now updated Figure 1c axis title to "Number of eruptions/250 years".

Reviewer #2 (Remarks to the Author):

I looked over the rebuttal and the changes to the manuscript in relation to my comments. I am happy with how they have addressed and rebutted my comments.

Reviewer #3 (Remarks to the Author):

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