Response letter to reviewer 2

We thank the reviewer for the provided feedback. Please find below our point-by-point responses (marked in blue).

Volcanic forcing is one of the most important inputs to modelling studies. Until recently it has been challenging to estimate it accurately even for recent centuries. Through a huge amount of work this paper and dataset now offers the chance to do it, albeit with large caveats and uncertainties, for the whole of the Holocene period. The dataset is based on painstaking synchronisation of records within Antarctica and Greenland, and between the two ice sheets. The data are used to assess S emissions in three crudely defined latitude bands. The output is then used to derive datasets of volcanic sulfur injection, and a latitude/time dataset of stratospheric aerosol optical depth.

I consider this a very useful and worthwhile dataset that certainly warrants publication in ESSD. The authors describe the steps they have taken and the weaknesses in a clear way. At the present moment this is the best dataset that could be produced, and the documentation of it is good.

The authors go quite a long way beyond the description of the data to discuss some implications of the data. This makes the paper extremely long and I fear some of their conclusions that deserve further discussion will be lost in what will be perceived as simply a presentation of the data – the authors may want to consider whether they want to take some of those points out into a separate paper, although I certainly would not insist on it. There are some aspects of the latitudinal classification that I find a little puzzling and that I will query in the detailed comments that follow. However, provided the authors consider the (mainly very minor) points listed below then I believe the paper could be published with minor revision.

Reply: We would like to thank the reviewer for their thorough and critical review of the manuscript. We have addressed the reviewer's comments one by one below and revised our paper based on their suggestions. We have reviewed other publications in ESSD and we have noticed that there is a wide range in the content of papers, from very brief descriptions of data to their interpretation. We are convinced that the dataset itself will be of great use to the scientific community, but at the same time we wanted to take the opportunity to highlight some of the open research questions related to global volcanism and climate, albeit without the deserved in-depth coverage. While not every reader and data user will need the details of our analysis and discussion points, they are intended to stimulate future research in the field and we would therefore like to keep the paper as detailed as possible.

Specific comments:

Line 26 "follows the global distribution of landmasses". Taken out of context in the abstract I think this leads the reader to think it means something much more detailed than it does. You only mean (as far as I can tell when it comes up later (line 741)) that the proportion of eruptions in the 3 boxes (NH, tropic, SH) is somehow related to the distribution of land between these 3 boxes. I think the reader seeing the abstract would think at the very least you were implying a relationship between eruption numbers/strength and the area of each continent. I'd suggest removing this from here or else spelling out the limited meaning you have.

Reply: Thanks for noting this. You are absolutely right. We did not intend to interpret anything beyond the obvious and purely qualitative correlation of the global distribution of known volcanic eruptions and our attributed latitudes of sulfur emissions. We will remove this in the abstract as suggested.

Line 67 East Antarctica is generally considered a place name and therefore East is capitalised.

Reply: We have corrected this.

Page 3. I appreciate they are mentioned later but I'm a little surprised that the potential of S isotopes to discriminate stratospheric and hence bipolar eruptions is not mentioned here.

Reply: We will add a sentence of the potential and limits of S-isotopes to discriminate between tropospheric and stratospheric sulfate formation and thus to attribute bipolar eruptions with some relevant references of recent studies (Burke et al., 2019; Crick et al., 2022). This is an emerging technique, which in the past had required large amounts of ice-core material (see Gautier et al., 2019).

Fig 2. Lower 2 panels. It's quite hard to discriminate the black and blue lines, I wonder if a different combination of colours would be better. This is quite a crucial figure to show that your methodology works so I think it needs to be presented in such a way that the synchronisation is really obvious.

Reply: We agree and we have changed the colors to improve readability. In addition, we will make available the necessary metadata (i.e. depths and ages for each correlated ice-core signal in the five ice-core records) as Supplementary Data. This allows assessment of the sequential number of annual layers (i.e. years) in between consecutive volcanic marker layers in the two independently annual-layer-counted chronologies of GICC05 (Greenland) and WD2014 (Antarctica).

Lines 194-6. Same sentence repeated.

Reply: We have corrected this.

Line 204. S is 32 g/mol, SO4 96 g/mol, NOT kg/mol!

Reply: We have corrected this.

Line 205. I'm not sure I understand the rationale for scaling the deposition at each site to that of WD. If you just want to give each record equal weight, then normalising the three records without scaling them would have worked. But does your method imply that somehow the deposition at WD is a "better" number than that at the other sites. I may have misunderstood the purpose, so it might just need a better explanation.

Reply: The linear scaling is used primarily to show in Figure 3 the relative amplitudes of the volcanic anomalies in the three different ice cores on a y-axis with a similar range of values. This allowed us to judge the plausibility of the volcanic synchronization without getting distracted by the differences in absolute concentrations due to the different site characteristics (see Figure R1). In principle, it does not matter which ice core is considered as the base because in a later step (Lines 219-221) a linear regression is performed against a more comprehensive ice-core network for the Common Era. Normalizing (or standardizing) the records as proposed

by the reviewer would result in comparable results (see Figure R2), though with different units (z-scores, [0,1]) which we believe would be less intuitive in comparison with existing records from ice-core arrays in Antarctica shown in Figure 4. We have added better explanation to the text and a note that the choice of scaling (or not) versus alternative methods such as standardization and normalization does not significantly affect our reconstruction results.
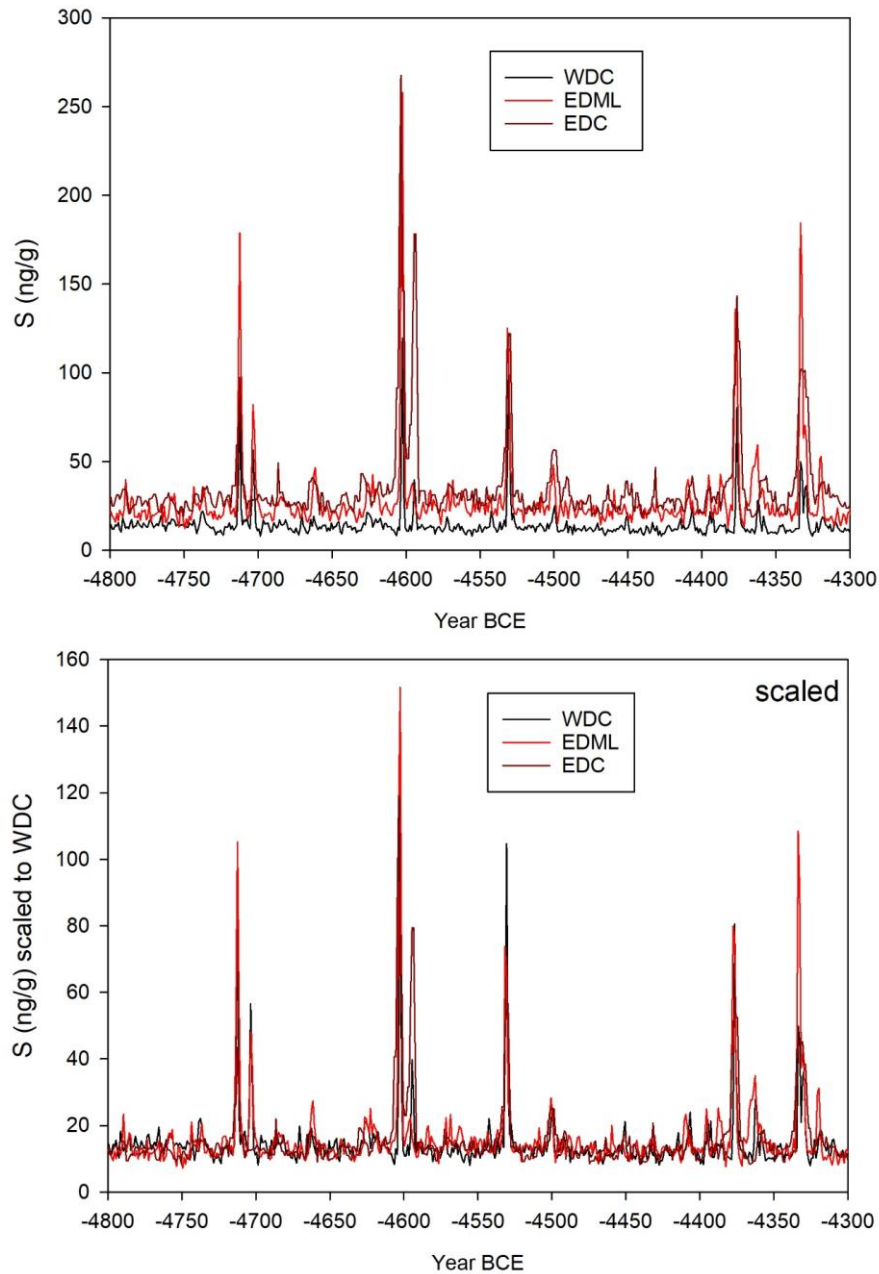
Figure R1: (Upper panel): Sulfur concentrations in the three ice cores from Antarctica for a representative time period 4800-4300 BCE. For the EDML and EDC ice cores, sulfur concentrations are derived from sulfate concentration data using $[S] = [SO_4^{2-}]/2.996$. (Lower panel): EDML and EDC were scaled to WDC for better comparability of relative peak magnitudes with respect to the background – a criteria used to judge the plausibility of age synchronization.
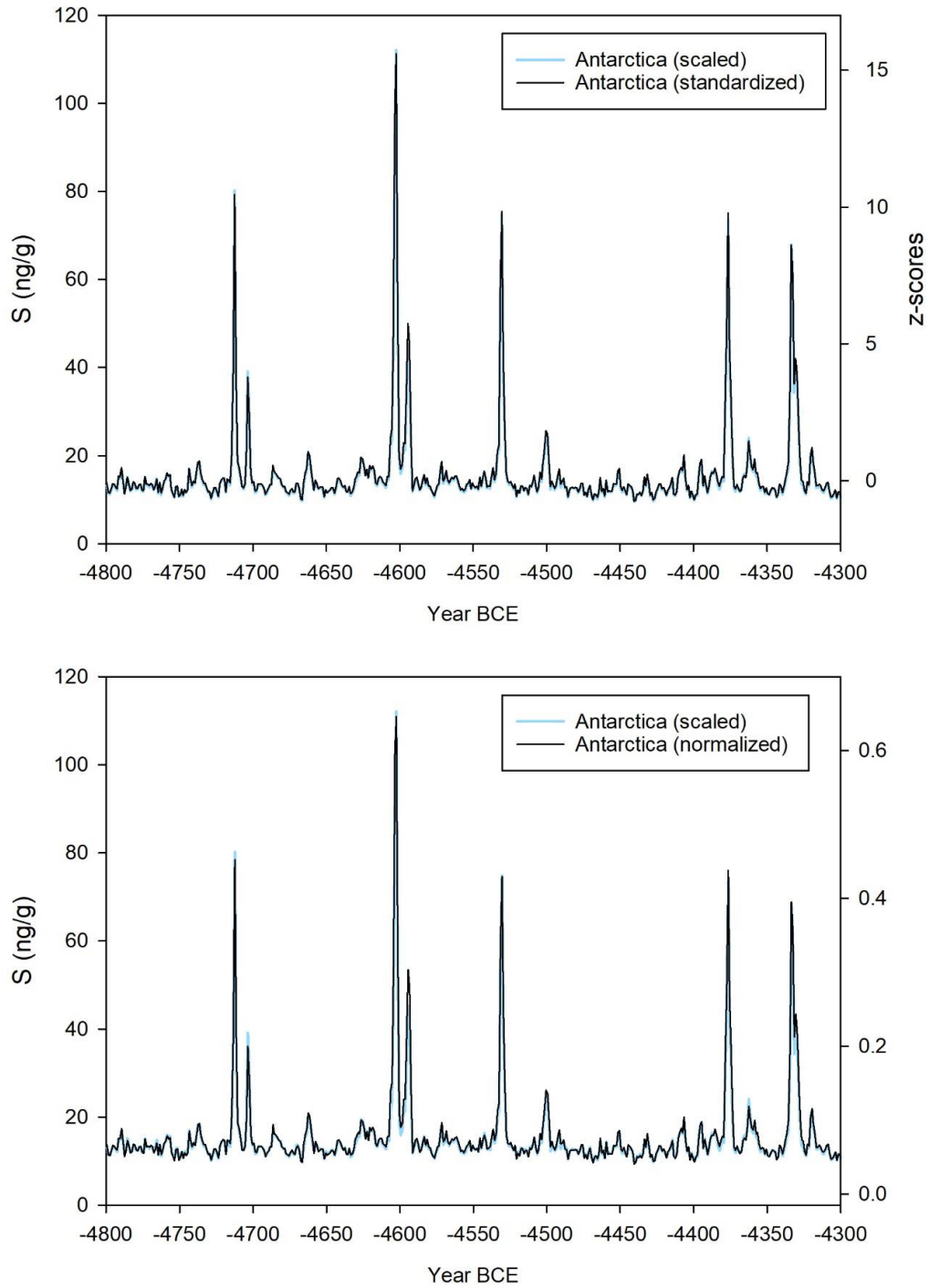
Figure R2: (Upper panel): Mean Antarctica (N=3) stacked sulfur concentrations (scaled to WDC, blue) versus stacked standardized Antarctica z-scores (black). (Lower panel) Mean Antarctica (N=3) stacked sulfur concentrations (scaled to WDC, blue) versus normalized concentrations (z-scores).

Line 207. I also think you should discuss the philosophy behind doing a stack as an average of the three (normalised) records. One could wonder whether the most accurate deposition rate is the average of the sites, or whether a missing peak at one site should be ignored and the best estimate is the maximum value at the three sites. I'm not arguing for that, but I think it's not self-evident (and therefore needs discussing) why the average is the best answer.

Reply: The reviewer raises here an important question that has also been addressed in the literature on several occasions (e.g. Gao et al., 2007, 2008; Crowley et al., 2012; Sigl et al., 2014). In this literature, reconstructions based on composite mean depositions to account for spatial variation in deposition are traditionally considered more reliable than those based on only a single ice core. Volcanic forcing for the Common Era reconstructed on the basis of composite records are widely used by climate model simulations and produce simulated temperature trends and magnitudes over the last millennium in very good agreement with climate proxy records. Nevertheless, there are site specific effects (e.g. such as redistribution by wind) which can obscure the sulfate records for individual events in various ways, or regional volcanic activity within Antarctica may equally produce differences in sulfate deposition among the ice cores. As discussed in Gautier et al. (2016), missing sulfate peaks following major volcanic eruptions are rare in Antarctic even in regions of low snowfall such as EDC in East Antarctica. Sigl et al., (2014) used ice-core records from EDC and EDML for the past 2 ka and detected volcanic sulfate in these ice cores for all eruptions with sulfate deposition in excess of that of Pinatubo 1991. For EDML and WDC a missing peak through snow loss is nearly impossible because of the high annual snow accumulation rates. In the revised paper, we will further underpin our choice of working with a stacked average with the relevant literature from above.

Line 213. Can you give a statistician's estimate of how many non-volcanic outliers you'd expect above 2xMAD?

Reply: I am afraid we can't provide such estimates because we don't have sufficient ground truth to test the skills of our detection threshold. It is worth noting that more conservative approaches have been taken (e.g. 4xMAD) for single ice cores (Traufetter 2004; Cole Dai 2021), which resulted in virtually no false positive volcanic detections but at the cost of missing many minor eruptions (false negatives) as revealed later when these single ice cores were assembled in a comprehensive array (Sigl et al., 2014). With our less conservative detection threshold and by using a stack approach, we reconstruct similar to Sigl et al. (2014) a more complete history of volcanic eruptions even if we may have potentially falsely attributed a few minor events which were not of volcanic origin. We believe our thresholds are well justified because common sulfate peaks over large spatial scales cannot readily be explained other sources (e.g. marine sulfate emissions are more localized). It remains however difficult to quantify the presence of false positive vs. false negative because 1) there are no independent sources of information on sulfate deposition over the ice sheets, 2) time periods with good knowledge of atmospheric volcanic sulfate loading (after 1979) are obscured in the ice cores from Greenland by anthropogenic sulfate pollution. We demonstrate that over the preindustrial period 1735-1900 CE we do not detect volcanic events in the GISP2 ice core (used for HolVol1.0) for which there wouldn't be support from four other ice-core records from

Greenland. Thus we are confident, that our choice of a 2xMAD threshold for GISP2 remains conservative enough to minimize false positives in the reconstruction Figure S3, [Supplement].

Line 265. Why have you used 1.5xMAD for Antarctica but 2xMAD for Greenland?

Reply: Site-specific and non-volcanic sulfate variability has been largely reduced by stacking in Antarctica which allows the use of a less conservative lower threshold for detection than in the GISP2 ice core. The same threshold of 1.5xMAD was used for the Common Era stack shown in Figure 4. Detailed discussion can also be found in the SOM of Sigl et al. (2014), in particular in Figure S8. We will add a justification and references for the choices of the thresholds in the revised paper.

Fig 4d: the colours on the plot in the pdf don't look like the ones named in the caption. In place of red and dark red, I see orange and brown (and am not sure which is which).

Reply: We updated the caption.

Line 245. I wondered why you only used GISP2 for Greenland when you obviously could in theory have used NGRIP, NEEM and GRIP (DEP) in at least part of the record.

Reply: One of our motivations was to use a consistent database of predictors throughout the Holocene to avoid introducing artificial trends that may arise by incorporating different measurements (e.g., DEP) for different ice cores at different times. Moreover, the four ice core records we used are the only publicly available records of sulfur or sulfate that span the Holocene. No sulfate records are publicly available for the early Holocene in NGRIP and NEEM – we look forward to when these data are available following the recent publication by Lin et al., (2022).

Section 2.5, line 328. I realise you need to choose something, but I found this assignment of 48°N, 37°S and 5°N to all unidentified eruptions a bit abrupt. Why these latitudes. And given that I would have assumed that a very high proportion of short-lived eruptions in Greenland were from Iceland, and from other Arctic locations, so why has 48N been chosen. I just think you need to be much more explicit about what a huge simplification this is, even if inevitable.

Reply: We would very much like to be able to attribute all these eruptions to specific locations, or at least latitudes. To your first point: We will specify how we attributed the default source latitudes for our database. We have collated from the Global Volcanism Program, 2013. Volcanoes of the World, v. 4.10.5. Venzke, E (ed.). Smithsonian Institution, database all volcanic eruptions during the Holocene with VEI≥4, which is thought to be the minimum detectable size in polar ice cores. We then calculated the mean latitude for all NH extratropical eruptions (>23.5°N) in this list; all tropical eruptions and all SH extratropical eruptions (>23.5°S); which resulted in 48°N, 5°N and 37°S respectively for our three broad latitudinal bands. In addition, we attributed sulfate signals of long (≥10 years) duration typical for eruption types (e.g. fissure eruptions) frequent in Iceland to the latitude of Iceland. Of course, many eruptions we currently attribute by default to 48°N may actually be from Iceland, though many others may also be from Kamchatka, the Aleutian Islands and or North America and well below the Arctic Circle (in the 50s°N). Many historic eruptions from Iceland also were sulfur-poor (e.g. Hekla 1104, 1158, Öræfajökull 1362) and strong westerlies limit sulfate deposition

towards Greenland to some extent. The bottom line is that we should not assume that Holocene NH signals in Greenland ice cores are dominated by Icelandic eruptions.

We also note that in the construction of aerosol optical properties and radiative forcing using the EVA tool, only the broad region of the eruption site is important (i.e., tropics, NH extratropics or SH extratropics), the exact latitude has no impact on the aerosol properties. When sulfur emissions are directly used in aerosol-climate models, differences in aerosol evolution depending on the latitude of the eruption within these broad regions may be relevant (see Toohey et al., 2019; Marshall et al., 2021), and our choice of the mean latitudes helps to minimize any potential bias in the long-term mean radiative forcing.

With no diagnostic tracer yet established to confidently link sulfate spikes to specific sources without tephra fingerprints, here we stick to our latitudinal attribution, but note that developing better constrains on source latitudes remains a future goal.

Line 490 and sup fig S4. You identify that there are more eruptions in the early Holocene, and later suggest this is mainly due to ice unloading. I can accept this, but surely that would only affect the high northern latitude source whereas it looks to me in Fig S4 and Fig S5 as if the tropics might also show a difference. But this is illogical as ice unloading will have had no effect at all, and indeed I think elsewhere you may say that there is no change in the rate of Antarctic S deposition (it certainly says that in the Lin et al paper which includes the lead author of this paper, in CPD). Should this be a concern?

Reply: We will clarify this section. There is strong evidence in particular from Greenland ice cores and other regional eruptions records (e.g. from Iceland) that the early Holocene was volcanically more active and this is most commonly attributed to rapid deglaciation. HolVol 1.0 seems to support this specifically with respect to long lasting volcanic episodes (that we attributed to Iceland) which clearly were more abundant in 9500-7000 BCE (Figure 5) and before that (Lin et al., 2022). However, there is an apparent increased activity also for bipolar eruptions which we have attributed to the tropics and this obviously cannot be explained by ice unloading. We can think of two hypotheses to explain this.

1) The increased volcanic activity in the tropics is real; it is unrelated to the unloading of ice-sheets and is within the range of other Holocene periods of increased activity in the tropics (e.g. the 13th century CE showed increased frequency of tropical eruptions).

2) The apparent increased volcanic activity may also be an artifact from our applied source attribution. In this scenario, some of the increased activity actually took place in glaciated NH regions (e.g. Iceland, Alaska, Kamchatka), but contemporaneous volcanic sulfate detected in Antarctica had caused us to attribute incorrectly the eruption source to the tropics. In consequence, we would have overestimated the true frequency for tropical eruptions and underestimated the true frequency of extra-tropical NH eruptions (some of which plausibly originated from volcanic areas experiencing ice unloading). We tested the plausibility of this scenario by comparing the distribution of asymmetry ratios (defined as the volcanic sulfate deposition in Greenland divided by the sum sulfate volcanic deposition in Greenland plus that for Antarctica) for all bipolar eruptions in HolVol1.0 from 9500-7000 BCE against, (1) all bipolar eruptions 7000 BCE-1900 CE and (2) against those of known historical tropical eruptions (e.g. Tambora, Krakatao,

Samalas) using the HolVol1.0 and eVolv2k databases. The results are shown in Figure A1, which we will add as an appendix to the revised manuscript. We find that the mean asymmetry ratio for attributed tropical eruptions between 9500-7000 BCE (N=98) are significantly (p<0.05) different (i.e. in our case indicating a stronger asymmetry of sulfate burden towards Greenland) from the mean asymmetry ratio for attributed tropical eruptions between 7000 BCE - 1900 CE (N=218). We interpret this result as an indication that the former group (during deglaciation) contains more eruptions that occurred further north than the latter group. The mean asymmetry ratio of both these groups of bipolar eruptions are significantly (p<0.01) different (i.e. again indicating a stronger asymmetry of sulfate burden towards Greenland) from the mean asymmetry ratios calculated for the limited number (N=11) of known tropical volcanic eruptions in ice core records in both HolVol1.0 and evolv2k.
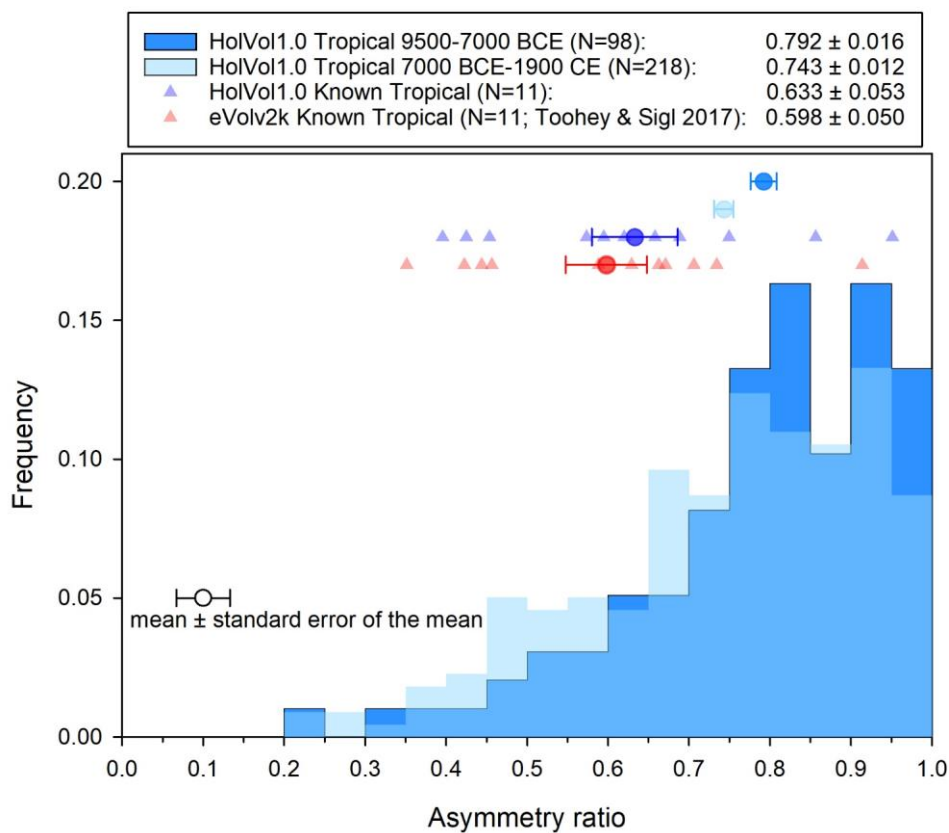
Figure A1: Asymmetry ratios [$D_{SO4\_GRL}/(D_{SO4\_GRL} + D_{SO4\_ANT})$] in HolVol1.0 and eVolv2k for attributed tropical eruptions and eleven known tropical eruptions detected in ice cores, where D is the mean sulfate deposition over Greenland (GRL) and Antarctica (ANT).

Altogether, we hypothesize that it is possible that our traditional assignment of bipolar signals to tropical source regions may be incorrect in some cases, disregarding the interhemispheric transport of aerosols from the extra-tropics to the polar regions of the opposite hemisphere. Especially during periods of exceptional activity in the northern latitudes (as during the deglaciation), this would lead to a seemingly increased rate of tropical eruptions, as it appears in HolVol1.0. Proving these hypotheses is extremely difficult, and really only possible through tephra identification of the sources in ice cores. Experiments with climate-aerosol models show in principle the possibility of interhemispheric sulfate transport also for non-tropical eruptions

(e.g., Toohey et al., 2019), but these depend strongly on season, plume height and other poorly constrained parameters and there is little agreement between different aerosol models even for well characterized historic eruptions.

We will add a small paragraph outlining these hypotheses with relevant literature and the new figure (in appendix). It is important to note that the attribution of volcanic signals to specific source latitudes carry large uncertainties, which may not be of great relevance for transient climate model simulations but must be considered when discussing spatio-temporal trends of volcanic activity in the context of a deglaciation-volcanism nexus.

Line 650 and surrounds. Related to the last comment, please be very careful here. Post-deglaciation increases of subaerial volcanism can indeed be imagined to be a feedback on CO_2, but if it's only the volcanoes in recently deglaciated areas, then it will only be a small proportion of the total volcanic emissions that are affected. It's therefore important to state that this will not apply to the (generally much more numerous and larger) tropical eruptions (why my question about Fig S4 and S5 is important).

Reply: The reviewer raises a very good point. We added a sentence putting the limited number of potential additional point sources of CO_2 into context with global-scale volcanic activity citing recent initiatives to inventory these emissions (Fischer et al., 2019).

Also please be careful to put the number from Ilyinskaya et al (which is not in the ref list) in context – the paper estimates that this one volcano "contributes up to 4% of global emissions from nonerupting volcanoes". The total volcanic emissions from all volcanoes are estimated (I believe) at 0.3-0.6 Gt/yr, about 1% of human emissions. I just think it's important the reader is not left with the impression such volcanoes could be a really significant and variable source of CO2 to the atmosphere as I do not believe this is correct in the context of human emissions.

Reply: Thanks for noting this. We have corrected the statement from the Ilyinskaya et al. (2018) reference and added the reference in the bibliography. We have also added the most recent observational assessments of the global present-day share of volcanic CO_2 emissions relative to anthropogenic emissions (Le Quéré et al., 2018). Volcanic CO_2 emissions to the atmosphere are somewhat constrained by the ice-core records of atmospheric CO_2. It is not the subject of this study, but with frequent volcanic episodes with durations in excess of 70 years revealed it is timely to evaluate the potential role of volcanic emissions of greenhouse gases (e.g. CO_2, water vapor) in the paleo-atmosphere.

Section 4.2. I didn't really know what you were trying to say in this section. Is it necessary?

Reply: We will revise this chapter to make the main message clearer to the reader. The key point is that tropospheric aerosols are the main source of uncertainty in studies of climate sensitivity over the last 100 years, and a major forcing on all CMIP6 simulations. However, over the Holocene and the last 2000 years, these are not considered or assumed to be constant. Our reconstruction now shows for the first time that tropospheric sulfate aerosols had strong multi-decadal emission variations. In order to understand any effects on global and regional climate, these must become more quantifiable in the future, which is what this paragraph is intended to encourage the research community to do.

Line 778-780, maybe mention S isotopes here as well.

Reply: Done.

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