Underestimated passive volcanic sulfur degassing implies overestimated anthropogenic aerosol forcing

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Key Points:

- Sulfur isotopes in a Greenland ice core show that passive volcanic degassing contributes 66% of preindustrial Arctic sulfate
- The volcanic inventory used by most climate models underestimates passive degassing, possibly due to missing hydrogen sulfide emissions
- Elevated preindustrial passive volcanic degassing reduces the estimated cooling effect of anthropogenic sulfate in the Arctic

Abstract

The Arctic is warming at almost four times the global rate. Cooling caused by anthropogenic aerosols has been estimated to offset sixty percent of greenhouse-gas-induced Arctic warming, but the contribution of aerosols to radiative forcing (RF) represents the largest uncertainty in estimating total RF, largely due to unknown preindustrial aerosol abundance. Here, sulfur isotope measurements in a Greenland ice core show that passive volcanic degassing contributes up to 66 ± 10% of preindustrial ice core sulfate in years without major eruptions. A state-of-the-art model indicates passive volcanic sulfur emissions influencing the Arctic are underestimated by up to a factor of three, possibly because many volcanic inventories do not include hydrogen sulfide emissions. Higher preindustrial volcanic sulfur emissions reduce modeled anthropogenic Arctic aerosol cooling by up to a factor of two (+0.11 to +0.29 W m^{-2}), suggesting that underestimating passive volcanic sulfur emissions has significant implications for anthropogenic-induced Arctic climate change.

Plain Language Summary

Sulfate aerosols are particles in the atmosphere that have a net cooling effect on the climate. One of the most uncertain aspects of climate modeling is the abundance of sulfate aerosols during the preindustrial era. Without knowing the amount of sulfate aerosols during the preindustrial, it is difficult to estimate how much anthropogenic sulfate aerosols have offset warming from anthropogenic greenhouse gases. In this study, we examine preindustrial sulfate aerosols in a Greenland ice core. We find that sulfate aerosols from passive (i.e. non-eruptive) volcanic degassing contribute almost two thirds of preindustrial Arctic sulfate aerosols in years without major volcanic eruptions. We compare this result to a state-of-the-art global model and find that most climate models use a volcanic emissions inventory that underestimates preindustrial passive volcanic sulfur emissions. That volcanic inventory only includes one type of sulfur emission (sulfur dioxide), but studies have shown that volcanoes emit hydrogen sulfide, which can also form sulfate aerosols. We show that higher emissions of volcanic sulfur during the preindustrial era decrease the estimated cooling effect of anthropogenic aerosols during the industrial era. Thus, the underestimate of preindustrial volcanic emissions in current climate models has significant implications for anthropogenic climate change in the Arctic.

1 Introduction

Anthropogenic aerosols have a net cooling effect on global climate and partially offset warming from greenhouse gases, but represent the largest uncertainty in estimating total anthropogenic radiative forcing (RF) from 1850-2019 (Szopa et al., 2021). Aerosol RF results from aerosol-radiation interactions (RF_{ari}), including scattering solar radiation (Twomey, 1967), and aerosol-cloud interactions (RF_{aci}), including changing cloud albedo (Twomey, 1977). Other aerosol effects such as impacts on cloud fraction and lifetime are uncertain, but may be significant in the Arctic (Shindell et al., 2013). Sulfate aerosols have the largest cooling effect of any aerosol and their contribution to RF also has the largest uncertainty (Szopa et al., 2021).

The magnitude of aerosol RF depends on preindustrial aerosol abundance due to the nonlinear relationship between aerosols and cloud albedo: as aerosol abundance increases, cloud sensitivity to aerosol decreases. Thus, one of the largest sources of uncertainty in aerosol RF is poorly constrained natural emissions of aerosol precursors (Carslaw et al., 2013; Gettelman,

2015), especially emissions of volcanic sulfur dioxide (SO_2) and marine dimethyl sulfide (DMS), which are dominant natural sources of Arctic sulfate aerosol (Abbatt et al., 2019; Legrand et al., 1997; Patris et al., 2002; Wasiuta et al., 2006). Other potential sources of sulfate aerosol, including carbonyl sulfide, dust, and biomass burning, are negligible in the Arctic (Abbatt et al., 2019; Kjellström, 1998; Legrand et al., 1997; Patris et al., 2002; Wasiuta et al., 2006). Although volcanic eruptions garner more attention in the climate literature, passive emissions of SO_2 are currently estimated to be about ten times the typical annual emissions of SO_2 from eruptions (Carn et al., 2017).

Satellites provide global daily observations of volcanic SO_2 emissions from eruptive and passive degassing (Carn et al., 2015, 2017). Ground-based observations show that these satellite observations provide a lower-end estimate on volcanic SO_2 emissions because satellite detection limits are too high to reliably detect passive emissions from weakly degassing volcanoes (Fischer et al., 2019). Furthermore, volcanic SO_2 emissions inventories are primarily derived from UV satellite measurements, which have data gaps at high latitudes in the winter months (Carn et al., 2017). In addition to underestimating SO_2 emissions, these inventories exclude emissions of other sulfur species such as hydrogen sulfide (H_2S), which is difficult to measure from space due to a lack of characteristic absorption bands in the near UV and an overlap of IR absorption bands with those of water vapor (Clarisse et al., 2011). Estimates of volcanic H_2S emissions range from 1 to 35 Tg S yr^{-1} (Halmer et al., 2002); the upper end of this range is three times the estimated global annual mean eruptive plus passive volcanic SO_2 flux of 11-13 Tg S yr^{-1} (Carn et al., 2015, 2017). After emission, H_2S is oxidized to SO_2 on the timescale of 1-3 days (D'Alessandro et al., 2009; Kourtidis et al., 2008; Pham et al., 1995), by which point it is too dispersed to be detected by satellite. Thus, satellite observations underestimate volcanic sulfur emissions (Carn et al., 2017; Fischer et al., 2019), but the magnitude of the underestimate and the contribution of volcanic sulfur to the global sulfur burden remains unquantified.

2 Quantifying preindustrial sources of ice core sulfate

We quantify volcanic and DMS-derived biogenic contribution to preindustrial (1200 to 1850 C.E.) Arctic sulfate aerosols by measuring sulfate concentrations (SO_4^{2-}) and sulfur isotopic composition ($\delta^{34}S(SO_4^{2-})$) in ice core samples from Summit, Greenland (see Text S1 for details on measurement methods). We select samples from years without influence from large volcanic eruptions (Figure 1, Cole-Dai et al., 2013; Gautier et al., 2019).

To estimate the relative contribution of volcanic and DMS-derived biogenic sulfate to total ice core non-sea salt sulfate ($nssSO_4^{2-}$), we assume that $\delta^{34}S(nssSO_4^{2-})$ is a concentration-weighted average of the mean biogenic sulfate isotopic composition ($\delta^{34}S_{bio}$) and mean volcanic sulfate isotopic composition ($\delta^{34}S_{volc}$):

$$f_{bio} + f_{volc} = 1, \text{ and}$$
$$f_{bio}\,\delta^{34}S_{bio} + f_{volc}\,\delta^{34}S_{volc} = \delta^{34}S(nssSO_4^{2-}),$$

where f_{bio} is the fraction of DMS-derived biogenic sulfate and f_{volc} is the fraction of volcanic sulfate. $\delta^{34}S_{bio}$ is well constrained by measurements of sulfur isotopic composition of marine biogenic compounds at $\delta^{34}S_{bio} = +18.8 \pm 0.3$‰ (Table S1 and Figure S1). Observations of sulfate from an inland Antarctic ice core far from the marine biogenic source show $\delta^{34}S_{bio} = +18.6 \pm 0.9$‰ (Patris et al., 2000), suggesting minimal fractionation due to transport and oxidation of marine biogenic sulfur (Text S2).

We estimate $\delta^{34}S_{volc}$ using two methods. First, we estimate $\delta^{34}S_{volc}$ by applying a Monte Carlo routine to a Keeling Plot (Keeling, 1958; Keeling et al., 1989; Pataki et al., 2003) of the ice

core observations to determine $\delta^{34}S_{volc}$ = +4.1 ± 0.5 ‰ (Figure S2 and Table S3) using similar assumptions and methods as in Patris et al. (2000, 2002) (Text S2). Second, we use direct $\delta^{34}S$ observations of volcanic gas and ash from 367 measurements of volcanic $\delta^{34}S(H_2S)$, $\delta^{34}S(SO_4^{2-})$, $\delta^{34}S(SO_2)$, and $\delta^{34}S$(bulk S) from 38 volcanoes around the world (Table S2) to yield $\delta^{34}S_{volc}$ = +3.8 ± 0.7 ‰, where the standard error of the mean is determined using a bootstrapping method (Figure S1 and Text S2). Both estimates of $\delta^{34}S_{volc}$ (+4.1 ± 0.5 ‰ and +3.8 ± 0.7 ‰) result in similar values for mean ice core f_{volc} (66% and 64%, respectively; Text S3), but we focus on $\delta^{34}S_{volc}$ = +4.1 ± 0.5 ‰ because this value more likely represents a regional signature and also incorporates any fractionation effects on $\delta^{34}S_{volc}$ during transport to Summit.

Figure 1 shows ice core $\delta^{34}S(nssSO_4^{2-})$ and $nssSO_4^{2-}$ concentration between 1200 and 1850 C.E. The mean $\delta^{34}S(nssSO_4^{2-})$ is +9.2 ‰, indicating that the isotopically lighter volcanic sulfur contributes about twice as much on average as the biogenic sulfur source. Figure 2 shows that the mean volcanic sulfate concentration (19.1 ± 7.1 μg kg^{-1}) is 2.0 ± 1.7 times larger than the mean DMS-derived biogenic sulfate concentration (9.4 ± 3.0 μg kg^{-1}) and that the mean fraction of sulfate from volcanoes (f_{volc}) is 66 ± 10%. Using $\delta^{34}S_{volc}$ = +2.5 ‰ from a small number of observations from volcanoes near Greenland also yields a dominant contribution from volcanic sulfate (f_{volc} = 59%) (Text S3). We also consider how our estimate for f_{volc} is affected by including a continental source of sulfur (e.g. H_2S emissions from vegetation, salt marshes, tropical forests, soils, and wetlands) based on Watts (2000), which results in f_{volc} = 58 to 60% (Text S4). These numbers are similar to a previous estimate of f_{volc} = 57% from Legrand et al. (1997) in a Summit, Greenland ice core, which was estimated by subtracting an assumed DMS-derived contribution to ice core sulfate based on the summertime peak in $nssSO_4^{2-}$.

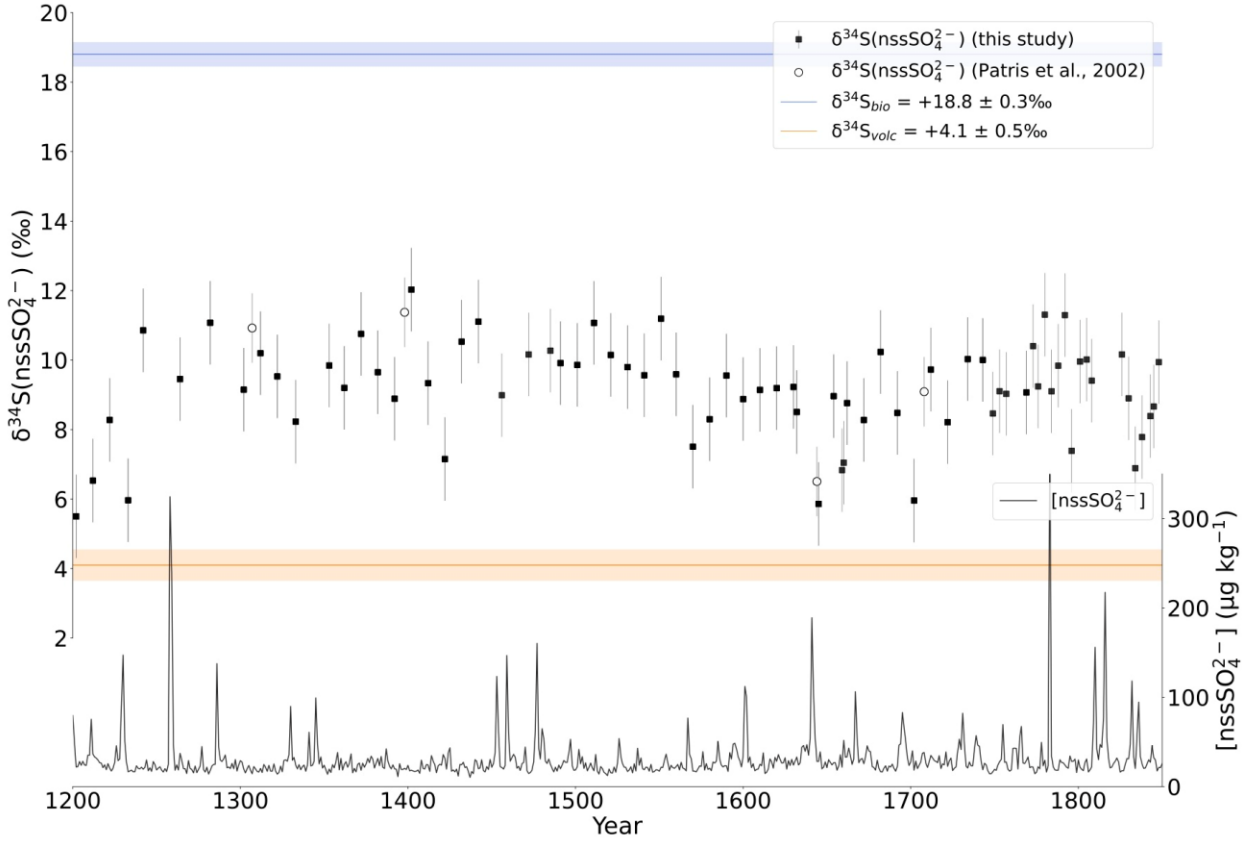

Figure 1. Decadal and sub-decadal ice core $\delta^{34}S(nssSO_4^{2-})$ (‰, black symbols) and annual mean $nssSO_4^{2-}$ concentration (μg kg^{-1}, gray line). Thick colored bars show the isotopic signatures of volcanic sulfur ($\delta^{34}S_{volc}$ = +4.1 ± 0.5 ‰) and DMS-derived biogenic sulfur ($\delta^{34}S_{bio}$ = +18.8 ± 0.3 ‰). The $\delta^{34}S(nssSO_4^{2-})$ samples were selected as one 2-year sample per decade from 1200-1750 C.E. and one 1-year sample every four years from 1750-1850 C.E. from years where $nssSO_4^{2-}$ was not influenced by large tropospheric or stratospheric eruptions (Cole-Dai et al., 2013; Gautier et al., 2019). Data from Patris et al. (2002) is also shown (circle). Error in $\delta^{34}S(nssSO_4^{2-})$ measurements is estimated based on replicate analysis of whole-process standards.

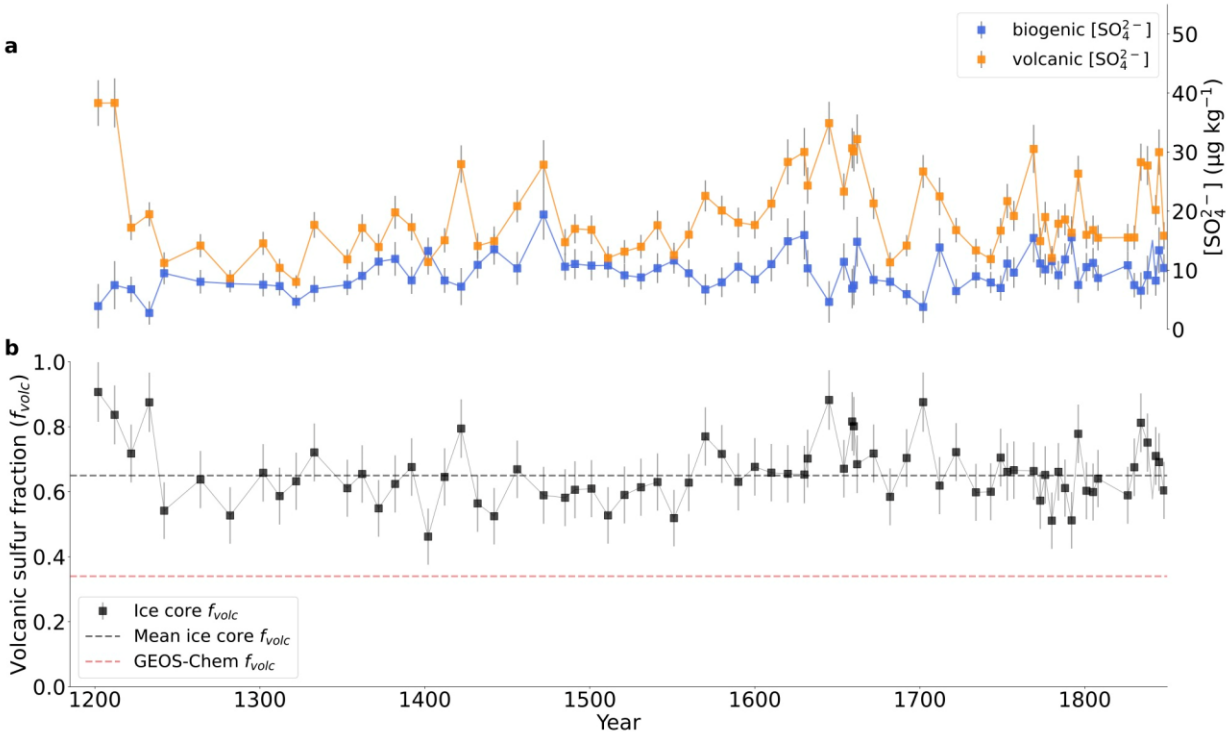

Figure 2. Volcanic and DMS-derived biogenic sulfate concentrations and volcanic fraction in ice core samples from preindustrial years (1200-1850 C.E.) without large volcanic eruptions. **(a)** Volcanic (orange) and DMS-derived biogenic (blue) sulfate concentrations (μg kg^{-1}) calculated with $\delta^{34}S_{volc}$ = +4.1 ± 0.5 ‰. **(b)** Volcanic fraction of ice core $nssSO_4^{2-}$ in each sample during the preindustrial (1200 to 1850 C.E.). Dashed gray line shows the mean volcanic fraction of ice core $nssSO_4^{2-}$ (f_{volc} = 66%). Dashed red line shows the GEOS-Chem simulated volcanic sulfur fraction (f_{volc} = 34%) in the air-mass source region of Summit with the default volcanic SO_2 emissions from Carn et al. (2015, 2017). Error bars were determined by propagating the uncertainty in isotopic source signatures and sample measurement error in both a and b.

3 Comparing ice core sulfate to a global model

To evaluate current estimates of the relative importance of volcanic and DMS-derived biogenic sulfate aerosol abundance in global models, we use the GEOS-Chem global 3-D chemical transport model (version 13.2.1, Text S5) described in Bey et al. (2001) driven by assimilated meteorology from MERRA-2. Volcanic SO_2 emissions are from Carn et al. (2015,

2017), updated annually in (Carn, 2022). The Carn et al. (2015, 2017) SO_2 emissions inventory is used in many global models and is the upper end of volcanic SO_2 emissions in Climate Model Intercomparison Project (CMIP6) models. The Carn et al. (2015, 2017) inventory includes passive and eruptive volcanic SO_2 emissions measured by the Ozone Monitoring Instrument (OMI) since 2005. Model DMS emissions are based on Lana et al. (2011). To simulate a preindustrial atmosphere, all anthropogenic emissions are turned off (Zhai et al., 2021). We use meteorology and volcanic SO_2 emissions from the year 2013, during which passive and eruptive volcanic SO_2 emissions in regions affecting the Arctic (i.e., Kamchatka, Alaska, and Iceland) were similar to the 2004-2017 median. To compare model results to ice-core derived estimate of f_{volc}, the average f_{volc} of the modeled tropospheric burden of SO_2 and sulfate is computed in the Summit, Greenland air-mass source region (120°W–30°E, 42°–90°N) based on the 5-day average aerosol lifetime in the Arctic and HYSPLIT backward trajectory analysis (Zhai et al., 2021). The modeled f_{volc} is similar when calculated with other methods, including f_{volc} of modeled sulfur deposition in the ice core region (Figure S3), and when modeled with meteorology from the year 2007 (Text S5).

Figure 3a shows that the modeled preindustrial f_{volc} over the Summit, Greenland air-mass source region using the SO_2 emissions reported by Carn et al. (2015, 2017) is 34%, which is 3.2 standard deviations lower than the mean ice core f_{volc}. The modeled volcanic sulfur contribution (f_{volc} = 34%) is lower than the observed f_{volc} in all 74 ice core samples representing 123 years between 1200 and 1850 C.E. (Figure 2b).

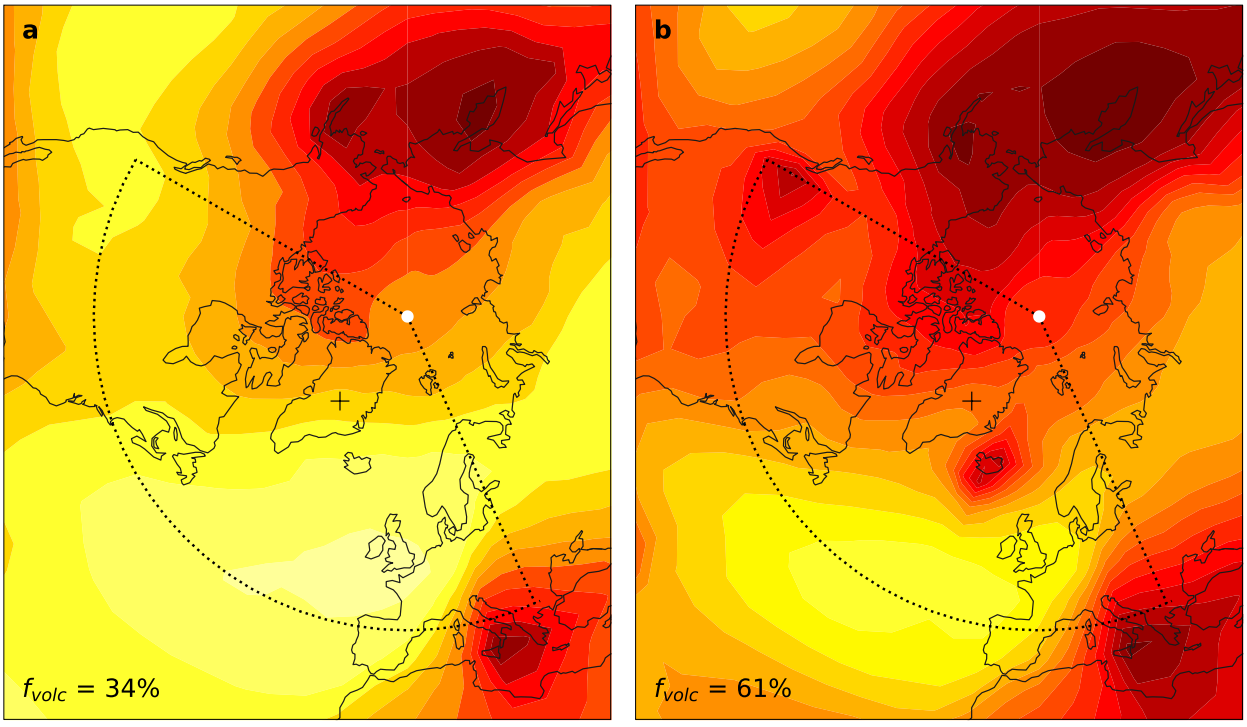

Figure 3. Modeled tropospheric f_{volc} in two preindustrial simulations. **(a)** Tropospheric f_{volc} in the preindustrial simulation with the default scenario volcanic emissions. **(b)** Tropospheric f_{volc} in the preindustrial simulation with emissions from the H_2S 1.7 scenario. Dotted black lines outline the 5-day back trajectory region (120˚W–30˚E, 42˚–90˚N) for the Summit, Greenland ice core

(location marked with "+") as described in Zhai et al. (2021). The mean f_{volc} of the sulfur (SO_2 + SO_4^{2-}) burden for the air-mass source region are shown in the bottom left.

4 Discussion

4.1 Biogenic and continental sulfur emissions and chemistry

A model underestimate in f_{volc} relative to the ice-core derived estimate f_{volc} could be explained by an overestimate in modeled DMS emissions or an underestimate in modeled volcanic emissions. Modeled preindustrial DMS emissions would have to be overestimated by a factor of three for DMS emissions to explain the model underestimate in f_{volc}. Ice core records show declining concentrations of methanesulfonic acid, an oxidation product of DMS, since the preindustrial (Osman et al., 2019), indicating that it is unlikely that present-day DMS flux is three times higher than that of the preindustrial. It is also unlikely that DMS emissions in the Arctic are overestimated by a factor of three; in fact, GEOS-Chem modeled atmospheric DMS concentrations are biased low in the Arctic (Text S6; Mungall et al., 2016). Although modeled DMS oxidation chemistry is simplified, uncertainty in modeled DMS chemistry cannot explain the discrepancy between modeled and observed f_{volc} (Text S6).

It is possible that other sources of sulfur not considered in global climate models could contribute to Arctic sulfate. For example, continental emissions of H_2S (e.g. vegetation, salt marshes, tropical forests, soils, and wetlands) contribute approximately 1.5 Tg S yr^{-1} globally (Watts, 2000). However, even if we assume this source contributes up to 4% of ice core sulfate, thereby lowering ice core f_{volc} to 58-60% from 66% (Text S4), the model would still underestimate f_{volc} in the Summit, Greenland back trajectory region.

4.2 Underestimate in passive volcanic degassing emissions

Given the low likelihood of an overestimation of DMS-derived sulfate in the Arctic, the discrepancy between modeled and ice core f_{volc} is best explained by an underestimate in passive volcanic sulfur emissions, which is consistent with comparisons between satellite and ground-based observations of SO_2 (Fischer et al., 2019) and the omission of volcanic H_2S emissions in models. It is also possible that preindustrial passive volcanic degassing was elevated relative to the present day. The sampled time period (1200-1850 C.E.) is during the "Little Ice Age" (LIA), usually defined as a period of relatively cool climate starting in the mid-thirteenth century and ending around 1850 C.E. (Grove, 2001). A driving factor in cooling observed during the LIA was an increased frequency of volcanic eruptions (Newhall et al., 2018). It is conceivable that passive volcanic degassing, which increases prior to and following volcanic eruptions (Carn et al., 2017), was also elevated during the preindustrial relative to the present day, which would exacerbate the underestimate in passive sulfur degassing emissions in the preindustrial. Elevated passive sulfur degassing around periods of increased eruption frequency has been suggested to explain differences between early and late 19th-century $\delta^{34}S(nssSO_4^{2-})$ in Antarctic ice cores (Takahashi et al., 2022).

To quantify and understand the factors contributing to the underestimate in preindustrial volcanic emissions, three volcanic emissions scenarios were prescribed in the model in place of the Carn et al. (2015, 2017) inventory: we label them the 371 scenario, the H_2S scenario, and the H_2S 1.7 scenario (summarized in Table S4). These emissions scenarios have increased passive

degassing of SO_2 relative to Carn et al. (2015, 2017) while leaving eruptive emissions unchanged. In the 371 scenario, volcanic SO_2 emissions are based on comparison between satellite and ground-based passive volcanic sulfur emissions from Fischer et al (2019) for volcanoes included in the Carn et al. (2015, 2017) SO_2 inventory. The 371 scenario also includes SO_2 fluxes from the 371 volcanoes identified as degassing by Fischer et al. (2019) that are not included in the Carn et al. (2015, 2017) SO_2 inventory. These 371 volcanoes are classified as either "hydrothermal" or "magmatic" and fluxes are assigned to be 3 or 7 t S day^{-1} (0.001 or 0.003 Tg S $year^{-1}$), respectively (Fischer et al., 2019). This includes 16 Icelandic volcanoes each emitting 3 t S day^{-1} (0.017 Tg S $year^{-1}$ in total). The 371 scenario results in a modest increase in f_{volc} in the Greenland air mass region from 34% in the default scenario to 36%, still three standard deviations below the mean ice core f_{volc} of 66% (Figure 2b).

In the H_2S scenario, we hypothesize that preindustrial and present-day simulations are missing a significant volcanic sulfur source due to the omission of H_2S from volcanic emissions inventories. Here, H_2S contribution is represented by increasing modeled SO_2 emissions due to the short 1- to 3-day lifetime of H_2S against oxidation to SO_2 (D'Alessandro et al., 2009; Kourtidis et al., 2008; Pham et al., 1995). Accordingly, SO_2 emissions are multiplied by a factor based on measured or predicted SO_2 to H_2S ratios (Halmer et al., 2002; Table S5). The H_2S scenario results in a f_{volc} of 46%, which is 2 standard deviations below the mean ice core f_{volc} of 66%.

In the H_2S 1.7 scenario, we multiply the SO_2 emissions from the H_2S scenario by 1.7 for each volcano. We choose the factor of 1.7 to approximate the mean ice core f_{volc} of 66%. As expected and shown in Fig. 3b, the H_2S 1.7 scenario produces f_{volc} of 61%, which approximately aligns with the mean ice core f_{volc} of 66%. This scenario implies that current estimates of preindustrial volcanic emissions are underestimated due to the omission of H_2S emissions and/or that passive volcanic degassing has decreased since the preindustrial.

We also consider a scenario where only Icelandic volcanoes have increased sulfur emissions and that these emissions are much larger than Icelandic emissions in the three aforementioned emissions scenarios (Text S7). SO_2 emissions from Iceland were 5.1 Tg S yr^{-1} in this scenario, which is 30 times larger than sulfur emissions from Iceland in the H_2S 1.7 scenario (0.16 Tg S yr^{-1}), but both scenarios are within the estimated range of sulfur emissions from Icelandic volcanoes based on observations from Icelandic hot springs (Text S7; Supplementary Data File 2). Icelandic volcanic sulfur emissions of this magnitude reconcile the discrepancy between the model and ice core (Text S7). Given the recent studies indicating that passive volcanic degassing CO_2 emissions in Iceland might be significantly underestimated (e.g., Ilyinskaya et al., 2018), it is possible that underestimated Icelandic volcanic sulfur emissions could explain most or all of the discrepancy between the ice core and modeled f_{volc}. This possibility also has significant radiative forcing implications (Text S7), and highlights the large uncertainty and disproportionate impact of Icelandic volcanic emissions in the North Atlantic and Greenland.

4.3 Radiative forcing implications

Models indicate that the cooling effect of anthropogenic aerosols would be lower than previously thought if the preindustrial sulfate aerosol abundance was higher because of the nonlinear relationship between aerosols and RF_{aci}: as preindustrial aerosol abundance increases, cloud albedo becomes less sensitive to anthropogenic aerosols (Carslaw et al., 2013; Gettelman et al., 2015). To explore the potential RF implications of our emissions scenarios, we consider three possibilities. First, we assume that the default volcanic emissions inventory from Carn et al.

(2015, 2017) accurately estimates present-day emissions, but underestimates passive volcanic sulfur emissions in the preindustrial. Second, we assume that passive volcanic sulfur emissions have not changed since the preindustrial, and that both preindustrial and present-day passive degassing emissions are underestimated. Overlapping volcanic and anthropogenic sulfur isotopic source signatures preclude quantifying volcanic sulfate in post-1850 ice core samples (Ghahremaninezhad et al., 2016; Patris et al., 2002; Wasiuta et al., 2006), therefore we cannot use post-1850 ice core measurements to evaluate this possibility. A third possibility is a combination of the first two: volcanic sulfur emissions in both present-day and preindustrial are underestimated and volcanic passive sulfur emissions were higher in the preindustrial relative to the present day. The RF implications of this third possibility will fall in between the first two.

To quantify the RF implications of an underestimate in passive volcanic degassing emissions in the preindustrial (first possibility) or both the preindustrial and present day (second possibility), we estimate RF for each possibility ($RF = RF_{ari} + RF_{aci}$), where RF_{ari} is calculated using GEOS-Chem (Text S5) and RF_{aci} is calculated using the simple heuristic model described by Wood 2021 (Text S8). We estimate ΔRF by subtracting RF with the Carn et al. (2015, 2017) inventory from RF with elevated passive degassing emissions representing the first or second possibility (Table S6). We quantify ΔRF for the first possibility by using the H_2S 1.7 scenario in the preindustrial and using the default scenario in the present day. We quantify ΔRF for the second possibility by using the H_2S 1.7 scenario in both the preindustrial and present day. We focus on the H_2S 1.7 scenario because it results in a preindustrial modeled f_{volc} (61%) approximately equal to the ice core f_{volc} (66%). The resulting ΔRF for both possibilities are summarized in Table 1. ΔRF ranges from +0.29 W m^{-2} (ΔRF_{ari} = +0.03 W m^{-2}, ΔRF_{aci} = +0.26 W m^{-2}; Table 1, Figure S4) where only preindustrial emissions are underestimated, to +0.11 W m^{-2} (ΔRF_{ari} = 0.0 W m^{-2}, ΔRF_{aci} = +0.11 W m^{-2}; Table 1, Figure S4) where both preindustrial and present-day emissions are underestimated. We estimate that underestimating Icelandic passive volcanic sulfur emissions could have an equally large or larger impact on radiative forcing (ΔRF = +0.55 W m^{-2}) (Text S7). This analysis neglects the effects of cloud adjustments to aerosol (i.e., impacts on cloud fraction, cloud lifetime, and semi-direct aerosol effects). Nevertheless, these calculations show that the impact of underestimating volcanic emissions on RF calculations is potentially large. Future studies using fully coupled atmosphere-ocean global climate models with enhanced volcanic emissions will be useful for more accurately quantifying RF implications and uncertainty.

Table 1. Radiative forcing (RF) estimates for different present-day and preindustrial volcanic scenarios.

Emissions Scenario	Volcanic Emissions Scenario in Present Day (f_{volc} of natural nssSO_4^{2-})	Volcanic Emissions Scenario in Preindustrial (f_{volc} of natural nssSO_4^{2-})	Arctic aerosol SW TOA RF [a] (RF_{ari} [b] + RF_{aci} [c]) between present day and in preindustrial (W m^{-2})	Difference between default Arctic RF and emissions scenario Arctic RF (W m^{-2}) ($\Delta RF_{ari} + \Delta RF_{aci}$)
Default	Default (f_{volc} = 34%)	Default (f_{volc} = 34%)	–0.55 (–0.10 + –0.45)	
Possibility 1	Default (f_{volc} = 34%)	H_2S 1.7 (f_{volc} = 61%)	–0.26 (–0.07 + –0.19)	+0.29 (0.03 + 0.26)
Possibility 2	H_2S 1.7 (f_{volc} = 61%)	H_2S 1.7 (f_{volc} = 61%)	–0.44 (–0.10 + –0.34)	+0.11 (0.00[d] + 0.11)

[a] The total shortwave (SW) top-of-atmosphere (TOA) radiative forcing (RF) is estimated as the sum of the RF from aerosol-radiation interactions (RF_{ari}) and RF from aerosol-cloud interaction (RF_{aci}).

[b] Difference between present-day radiative effect from aerosol-radiation interactions and preindustrial radiative effect from aerosol-radiation interactions.

[c] Estimated present-day RF from aerosol-cloud interactions (Text S8).

[d] ΔRF_{ari} between the default possibility and possibility 2 is negligible compared to ΔRF_{aci}

5 Conclusions

Our results indicate that passive volcanic degassing sulfur emissions influencing the Arctic are underestimated by up to a factor of three. We show that increased volcanic sulfur emissions from passive degassing results in estimated Arctic anthropogenic aerosol cooling that is up to a factor of two lower in magnitude. An overly strong anthropogenic aerosol cooling due to underestimated passive volcanic sulfur degassing could at least partially explain excessively strong aerosol cooling in CMIP6 climate models (Dittus et al., 2020) and the underestimates of modeled Arctic amplification compared to observations (Rantanen et al., 2022).

Quantifying passive volcanic sulfur degassing emissions is critical for constraining anthropogenic aerosol forcing. More observations of SO_2 emissions from passive volcanic degassing are required to constrain the magnitude of the underestimate in the passive volcanic SO_2 emissions inventory based on satellite measurements in Carn et al. (2015, 2017). Additionally, H_2S, which is typically neglected in volcanic emissions inventories used in global climate models, should be considered a potentially important contributor to the global atmospheric sulfur budget and thus climate.

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Open Research

All ice-core data and Iceland volcanic gas observations are available in the National Science Foundation (NSF) Arctic Data Center at https://doi.org/10.18739/A2N873162. GEOS-Chem version 13.2.1 code is publicly available at https://doi.org/10.5281/zenodo.5500717.

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