Quantifying southern high latitude sulfate sources using the spatial pattern of sulfur isotopes in Antarctica

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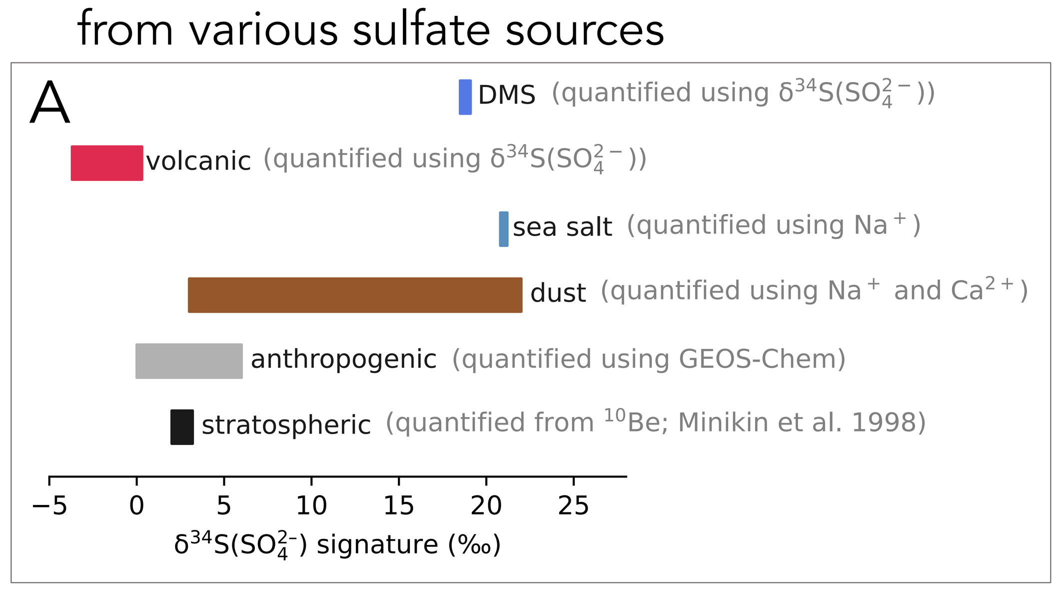
Motivation: Southern Ocean aerosols

- Uncertainty in aerosol radiative forcing motivates investigation of aerosols in the Southern Ocean region as a proxy for the preindustrial atmosphere.
- Sulfate is an important aerosol in this region, and models suggest phytoplankton dimethyl sulfide (DMS) is the largest source of sulfate during austral summer. However, sulfur isotopes of sulfate ($\delta^{34}S(SO_4^{2-})$) in Antarctic ice cores suggest that another source has significant influence.
- We use ice core sulfur isotopes and GEOS-Chem to ask: **What are the important sulfate sources in this region? How does this compare to existing assumptions?**

Methods: Ice cores and GEOS-Chem

1. Compile ice core measurements of $\delta^{34}S(SO_4^{2-})$ from the previous studies (see Figure 1b) and determine sources (see Figure 2):
2. Use GEOS-Chem 13.2.1 with several model configurations to determine relative contribution from various sulfate sources



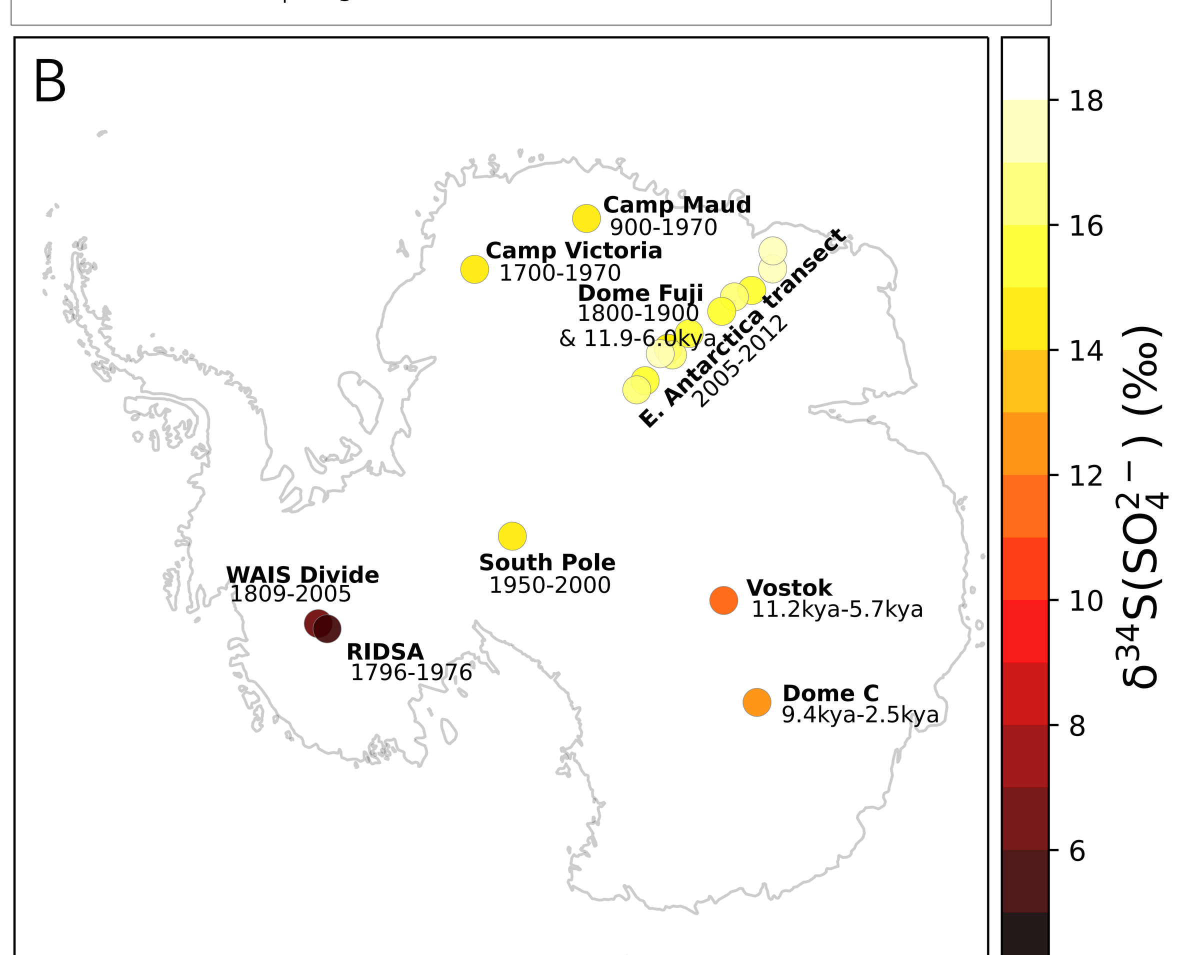


Figure 1. A) Sulfur isotopic signatures for each source of sulfate from previous studies. B) $\delta^{34}S(SO_4^{2-})$ in ice core and snow samples from Antarctica from previous studies: Alexander et al., (2003), Jonsell et al. (2005), Kunasek et al. (2010), Patris et al. (2000), Pruett et al. (2004), Takahashi et al. (2022), Uemura et al. (2016, 2022). Colored circles show the mean $\delta^{34}S(SO_4^{2-})$ at each location over the time period presented in each study. Volcanic eruptions are excluded when possible.

Ice core fraction of volcanic sulfate by region

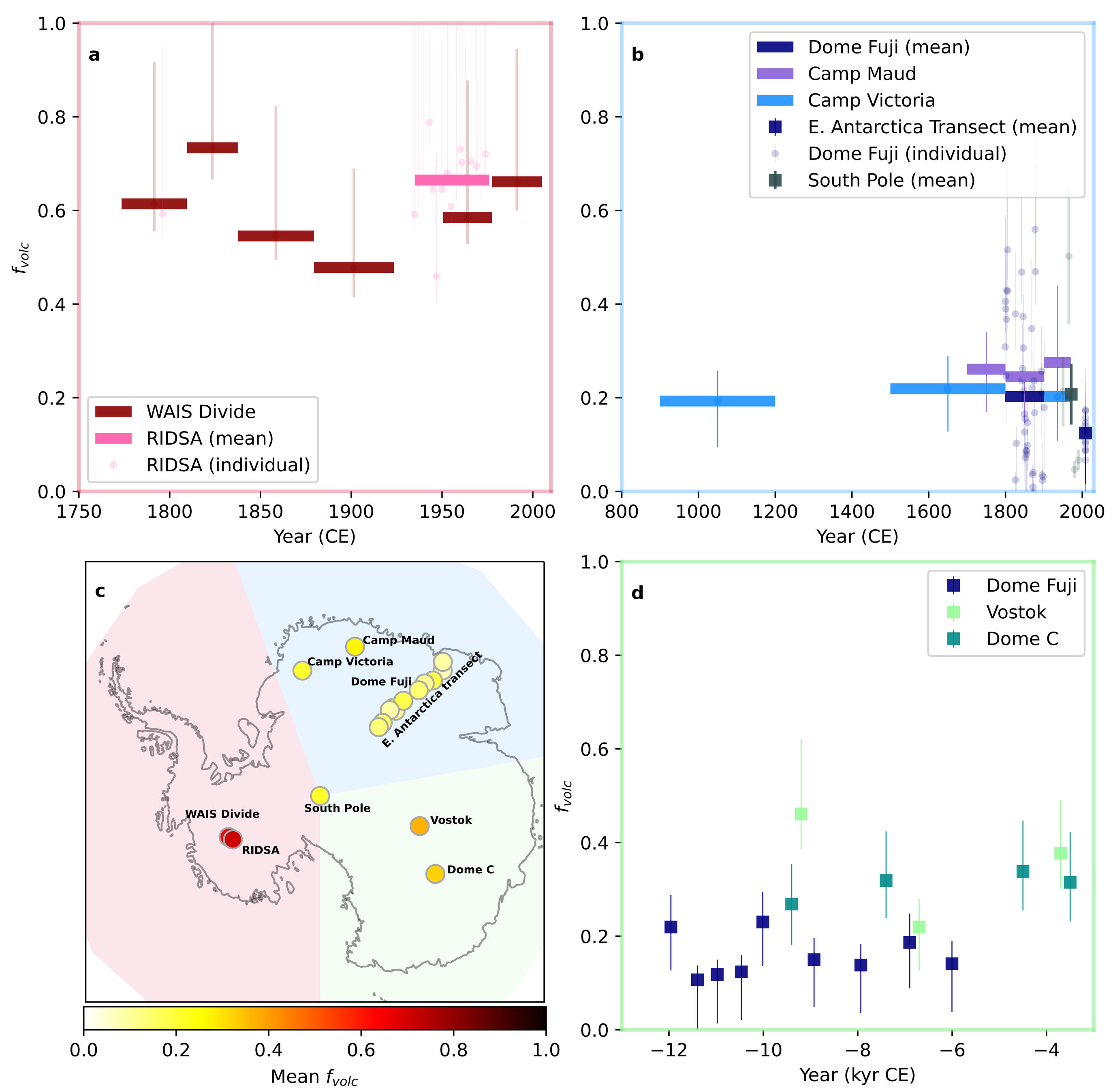


Figure 2. West Antarctic has a high volcanic sulfate fraction compared to East Antarctica. The volcanic sulfur fraction (f_{volc}) in **a)** West Antarctic ice core samples from WAIS Divide and RIDSA. **b)** East Antarctic ice core and snow samples from Queen Maud Land and the South Pole, and **d)** Wilkes Land in East Antarctica, including Dome C and Vostok, as well as Dome Fuji in Queen Maud Land. **c)** Spatial pattern of mean f_{volc} in Antarctica, with the land color corresponding to the subplot a, b, or d. Note the different x-axes on a, b, and d. Error bars show range of f_{volc} based on sensitivity tests. **West Antarctica has a higher f_{volc} than everywhere else. Why?**

Comparison with atmospheric DMS

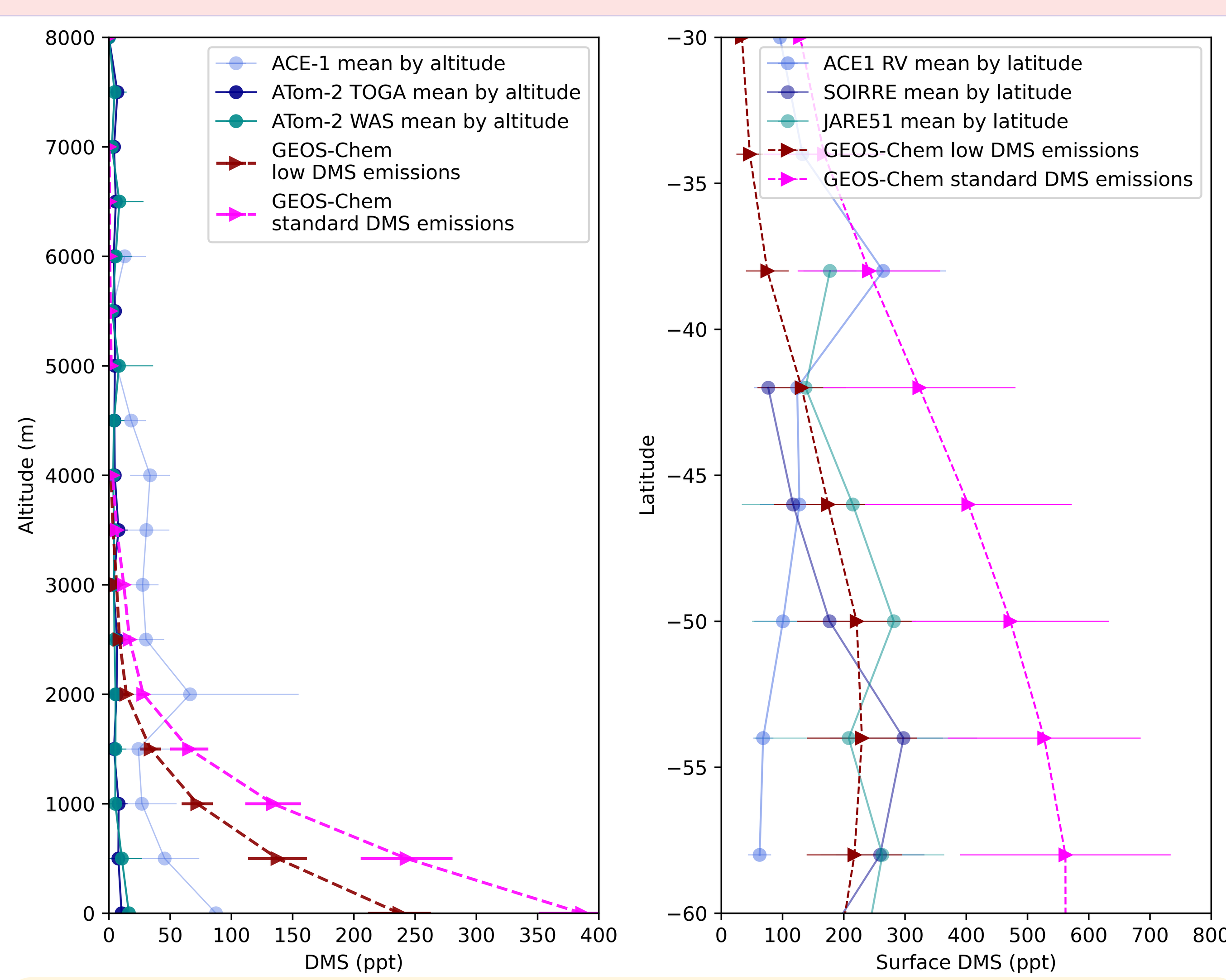


Figure 4. Comparison of DMS observations from aircraft (a) and ship (b) observations and GEOS-Chem simulations. GEOS-Chem simulations include the standard model with standard DMS emissions from Lana et al. (2011) and a sensitivity test with lower DMS emissions based on Bhatti et al. (2023). Observations are from aircraft (ACE-1, ATom-2) and ship (ACE1 RV, SOIREE, JARE51).

Model simulations varying DMS and passive volcanic degassing emissions

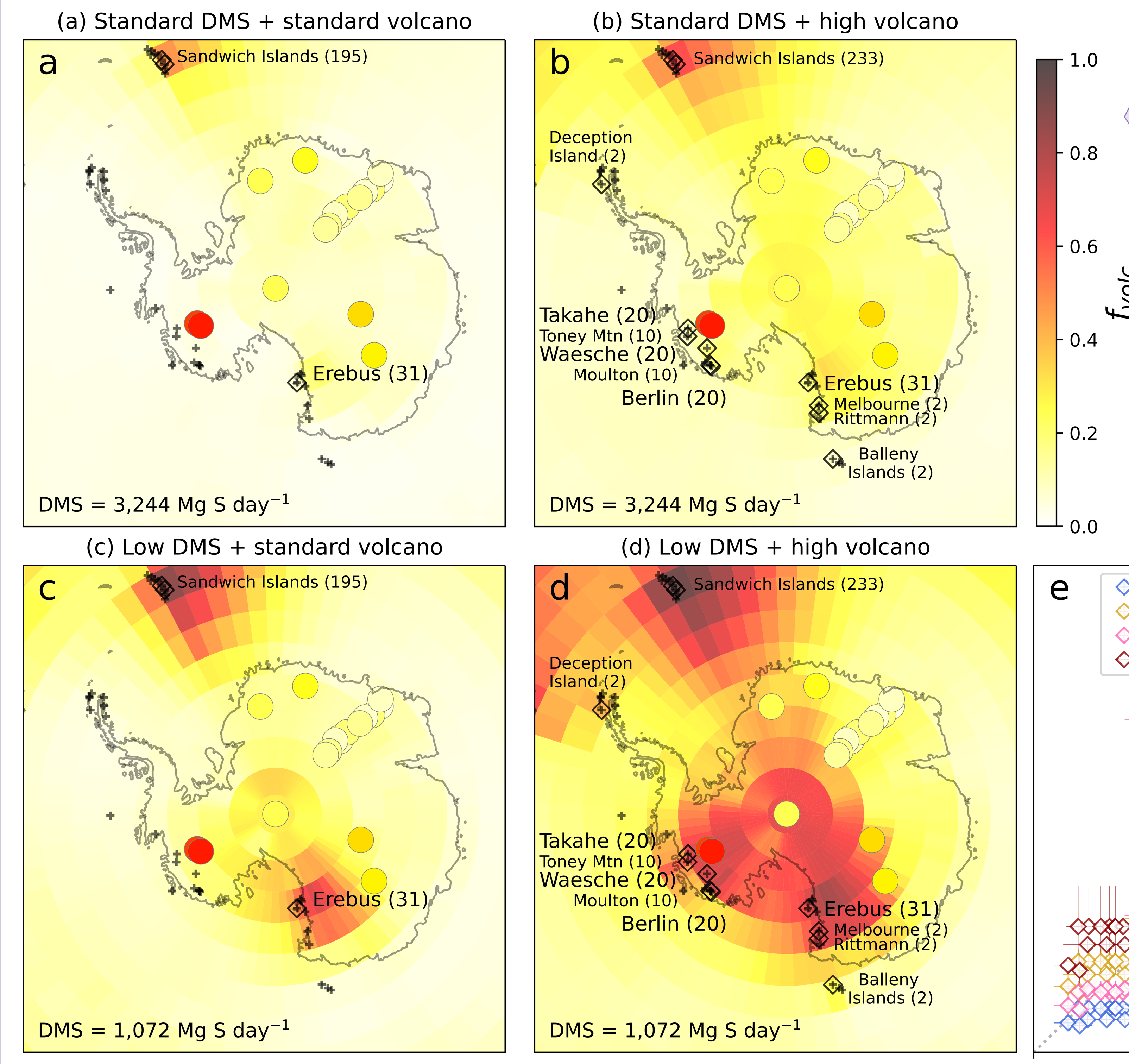


Figure 3a shows that in the standard model, f_{volc} ranges from 5–20% and underestimates observed f_{volc} in all 22 ice core locations. Sensitivity tests varied modeled meteorology, wet deposition scheme, and DMS oxidation chemistry, and could not reconcile this discrepancy.

In 3b, 3c, and 3d, we tested three emissions scenarios:
b) Standard DMS and high volcano
c) Low DMS and standard volcano
d) Low DMS and high volcano,
Where low DMS emissions are from Bhatti et al. (2023) and high volcano emissions are hypothesized H_2S emissions or past unobserved SO_2 emissions.

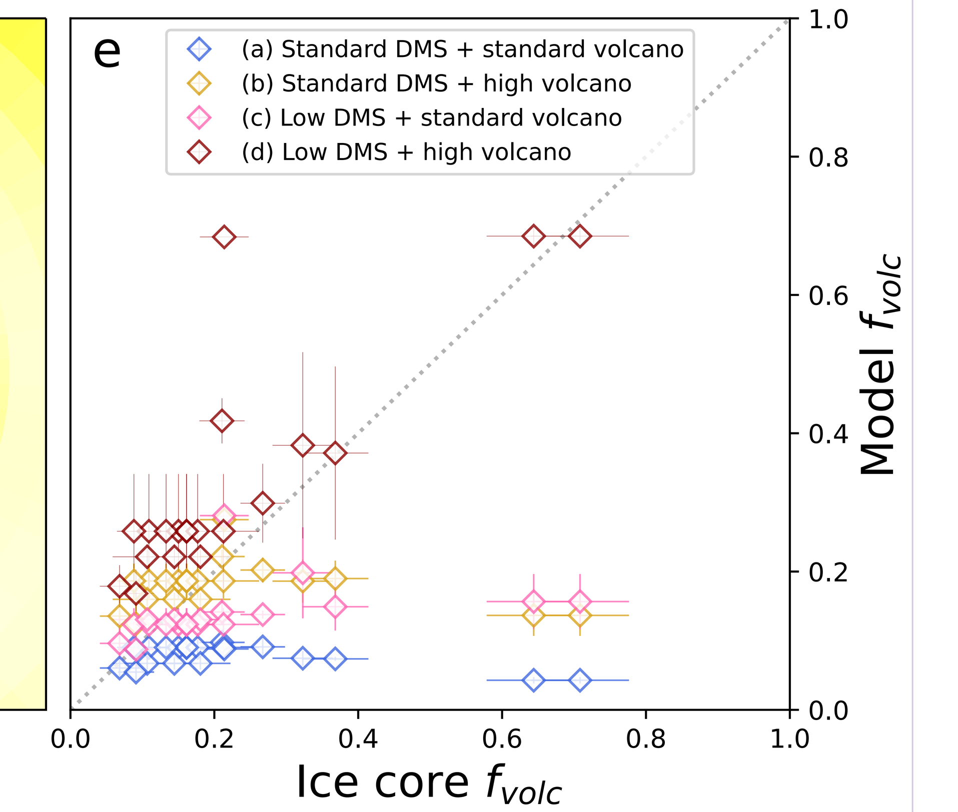


Figure 3. Lower Southern Ocean DMS emissions AND added West Antarctic passive volcanic degassing emissions are needed to explain spatial pattern of sulfur isotopes in Antarctica. Spatial pattern of volcanic sulfur fraction (f_{volc}) over different modeled DMS and volcanic emissions scenarios compared to ice core f_{volc} from Figure 2. The passive degassing emissions from each volcano are shown in parentheses next to each volcano name, and the annual DMS emissions south of 60°S are shown in the bottom left corner. Panel e shows comparison of ice core and modeled f_{volc} in the grid cell containing each ice core. Error bars in 3e show the range in f_{volc} in the grid cells immediately adjacent to the grid cell containing each ice core observation.

Conclusions

- Sulfur isotopes in Antarctic ice cores show that a high fraction (>50%) of West Antarctic sulfate and a substantial fraction (~20%) of East Antarctic sulfate comes from passive volcanic degassing
- The standard inventories used in GEOS-Chem cause the model to underestimate volcanic sulfate and/or overestimate DMS-derived sulfate in Antarctica
- Adding assumed passive volcanic degassing emissions in West Antarctica and implementing lower Southern Ocean DMS reconciles the discrepancy between model and observations
- Climate modelers should consider uncertainty in emissions from both DMS and volcanoes when studying aerosol-cloud interactions in the Southern Ocean region

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