

The authors have largely rejected the major concerns in my first-round review and have argued for the approaches and interpretations presented in their original manuscript. My main comments for the second round of review are as follows.

Upon reviewing the manuscript again, I am more convinced by the correlation between ssNa and COS at the same depths in the ice core. However, I am still not convinced that applying a correction based on this relationship is appropriate.

We regret that this disagreement persists. We insist that applying the ssNa correction is only appropriate course of action. An atmospheric interpretation without applying the correction would mean ignoring the strong evidence for the presence of COS production in the ice sheet. We will not include an interpretation to the manuscript that we do not believe is true.

The authors have assumed that this correlation is due to in situ production. I think this is one possible explanation, but I don't think it is the only possible explanation.

We cannot identify any plausible alternate explanation in the reviewer comments below.

In favor of the authors' assumption and approach is the observation that glacial-period and deglaciation COS value agreement between different ice cores is overall improved by this correction (with the caveat that the WD and TD ice core data seem even more complicated to interpret because of the hydrolysis correction that also has to be applied). However, there remain parts of the record where the different ice cores disagree (most notably around 19 ka and in the later part of the deglaciation). Further, the high scatter in glacial COS values (which was the main reason in situ production was postulated) does not seem to be improved by the correction in the scenario where there is no multi-point smoothing of the COS (Fig 4b, scenario G1).

The reviewer's statement about the high scatter in glacial period COS values being the main reason why in situ production is postulated is not accurate. The in situ production is postulated primarily as an explanation of the discrepancies between records from different sites, specifically during the last deglaciation, which is the most prominent climate event during the period our record covers. The detailed reasoning is presented on L267-297. Below, we present one sentence from this section (L287-289) to demonstrate this fact:

"Alternatively, the discrepancies between the records could be due to production in the ice sheet resulting in significant amounts of excess COS in glacial period ice; this possibility was not considered by Aydin et al. (2016)."

Given that the correction is not intended to correct the spikes, the persistence of spikes after the correction cannot be perceived as evidence of production not happening. We cannot even rule out the possibility that the spikes represent atmospheric fluctuations. We state very clearly in the manuscript that the applied correction does not address high frequency variability because the same depth relationship is driven primarily by millennial scale variability (section 3.1). In multiple analyses scenarios presented in section 3.1, we also clearly demonstrate that the existence of spikes does not impact the applied correction. We could assume the spikes were also a result of production, eliminate them from the record by determining a baseline following some examples in literature, then conduct the exact same analyses presented in the manuscript and arrive at the same results, including the same-depth relationships driven by

millennial scale variability, and the same corrections for production that happens in the firn. The only difference would be that the corrected glacial period COS levels would be somewhat lower because the spikes would have been eliminated from the record before averaging. In essence, our approach of not trying to eliminate the spikes from the record is the more conservative approach.

The manuscript text about the 19 ky feature has been revised based on another comment by the reviewer (see below). As we note in the reply to that comment, we do not perceive a contradiction between the main point of the manuscript about lower atmospheric COS during the last glacial period and the implications of this observation on ocean productivity and whether the 19 ky feature is a real atmospheric event or not. Note that the 19 ky feature is delineated by 4 measurements out of 574.

Finally, the fact that the glacial period record may include some artifacts caused by production even after the correction does not invalidate the applied correction as an appropriate method. It only means that the glacial period COS levels could in fact be somewhat lower than what we present in the paper.

Delta age for the South Pole ice core during the glacial (1500 – 2700) is in the same range as peak-trough age separation of AIM (Antarctic isotope maximum) events. Is it possible, for example, that higher ssNa at AIM peaks correlates (imperfectly) in depth with higher COS at AIM troughs?

No, this is not possible. This should be evident from the SPC14 ssNa record plotted versus the composite Antarctic CO₂ record in Fig 4c. In a more general sense, any property measured in ice that correlates with ssNa will correlate with same-depth COS. The possible environmental causes of the ssNa variations, whether or not AIM events are relevant in this context, do not have any bearing on the interpretation of the the same-depth relationship between ssNa and COS. The same-age anticorrelation between ssNa and COS that emerges after the correction can be interpreted as a climate driven relationship. We focus solely on the glacial/interglacial change in the interpretation to keep the interpretation section focused on the most prominent feature of the record.

Alternatively, is it possible that there are multiple COS-altering mechanisms in the ice core, and in situ production related to organic S impurities that are correlated with ssNa deposition is happening at the same time as COS destruction by another process?

Yes, there are multiple mechanisms that alter COS in the ice cores. In addition to the production process, which is the main theme of this manuscript, COS undergoes in situ hydrolysis loss in ice cores. This fact is clearly acknowledged numerous times with relevant citations, including as early as L49-50 in the introduction: “Previous measurements of COS in Antarctic ice cores revealed slow, temperature-dependent degradation of COS in the ice core air due to hydrolysis (Aydin et al., 2014).” At lower ice sheet temperatures, hydrolysis loss is very slow and can practically be ignored at the South Pole. However, the WAIS Divide and Taylor Dome ice cores require a hydrolysis loss correction as well as accounting for the production (L267-270): “There are two other ice core COS records that extend back to the last glacial period. They are from the Taylor Dome (TD) and the West Antarctic Ice Sheet Divide (WD), Antarctica (Fig. 1c). Both of

these sites are warmer than the South Pole, therefore the TD and WD measurements require a correction for temperature-dependent hydrolysis loss (Aydin et al., 2014; 2016).”

If the reviewer is referring to some other process, they are not offering any specific evidence supporting this idea. It is impossible for us to agree with or refute evidence that we do not see.

With regard to the inferred 2 – 4 times lower ocean COS source in the LGM as compared to the Holocene, I again think that this is a possible interpretation, but I am not convinced that this is the only possible interpretation. As the authors mention in their response, the evidence for ocean biological productivity changes during the deglaciation is mixed – there is paleoceanographic evidence for some regions being more productive during the LGM, and other regions less productive. Because of this, a scenario with no large ocean emission changes over the deglaciation (a scenario that would result if the in situ ssNa-based correction is not applied) seems possible to me.

We stated in the previous round of review that the glacial/interglacial change in global ocean productivity is an open science question, countering the reviewer’s statement that there was plenty of evidence productivity was not lower during the last glacial period. We present the atmospheric COS record as an important piece of evidence that supports globally lower ocean productivity. Regional changes in ocean productivity are not relevant in the context of atmospheric COS variability. As we stated in the previous round of the review, we offer an in depth discussion on this supported by plenty of citations (L527-596). Much like the first round of reviews, the reviewer does not directly challenge any specific evidence and arguments offered in the discussion section. If the reviewer chooses to believe ocean productivity does not change between glacial and interglacial climates, that is their prerogative. We do not intend to convince everyone with one paper.

Related to this point is the large inferred COS peak at the end of LGM (≈ 19 ka; COS on par with Holocene values), which is unexplained.

In the previous versions, we refrained from speculating about the nature of the positive excursion around 19 ky. We do not feel an obligation to offer an explanation for every feature in the record. Our inability to offer an explanation for any particular aspect of the record does not invalidate the explanations we offer for the data set as a whole.

That said, based on the current and the previous reviews, it appears that the reviewer is suggesting an interpretation of the 19 ky peak apparent in the SPC14 record as an atmospheric signal that may have resulted from an increase in ocean productivity. This is indeed possible, although we do not feel confident enough to make a strong claim about this without confirmation with measurements from other ice cores that there is indeed a peak at that time horizon. We would like to note that the current interpretation of lower biological productivity during the last glacial period does not in any way preclude the possibility of a relatively short-lived spike in ocean productivity during the LGM superimposed on the low baseline. We revised the relevant sections in the paper to clarify this and incorporate the reviewer’s suggestion into the manuscript (L515-524):

“The 19 ky peak is characterized by four wet COS measurements and coincides in time with a shorter-lived sharp peak in ssNa (Fig. 4c). Given the prominence of spikes in the glacial period COS, we suspect that at least one of the measurements characterizing the 19 ky COS peak, possibly the highest measurement dating older than 20 ky, may be a coincidental, non-atmospheric spike while the other three measurements may characterize an atmospheric excursion of 50-100 ppt that is closer in duration to the peak in ssNa. This may explain why we do not see this feature in the WD COS record. An atmospheric COS excursion of this magnitude would represent a sudden and significant departure from the biogeochemical balance that maintains the low atmospheric COS levels during the LGM. Based solely on the magnitude, it could only be caused by an increase in ocean sulfur gas emissions or a decline in land biosphere uptake since these are the two major natural components of the COS budget (Table 1). This feature warrants further investigation if replicated with high resolution measurements from different ice cores.”

Considering all of the above, my recommendation is still that the authors consider and include an alternative scenario in which the in situ correction is not applied.

As we stated in the beginning of our responses, we will not include an atmospheric interpretation of the uncorrected record, simply because we do not believe the measured COS values in the SPC14 ice core, particularly from the glacial period, represent atmospheric mixing ratios. We do not see any convincing evidence in the reviewer comments that suggests the opposite is true.

A more minor issue -- I am still not convinced that the in situ production the authors propose takes place mainly in the firn layer, for the reasons I highlighted in my original review. Wouldn't it also be possible, for example, that the concentration of impurities with depth that the authors mention would make production in deeper ice more likely than in shallow firn?

We are not sure what the reviewer means by concentration of impurities with depth. The impurity levels are higher in deeper ice from the last glacial period. This is not because impurities migrate in the ice sheet to deeper horizons after deposition. It is because surface snow and shallow firn were characterized by high impurity levels during the last glacial period.

But I agree with the authors that this process seems to be substrate-limited, otherwise there would a steady increase trend with depth as the authors suggest.

I think the authors have addressed my other comments sufficiently well.