Second Review of: "Drivers of Atmospheric Volatile Methylated Sulfur Variability Across the Southern Ocean and Antarctic Coast" by Mynard et al.

The authors have adequately addressed reviewer's concerns, though two points regarding the analytical chemistry require further clarification to strengthen the manuscript.

Referee Response 1 L95: The PTR was calibrated every 5 days. Can the authors provide a figure that shows that the analytical system was very stable and not more frequent calibrations were necessary?

Author Response 1: The initial aim was to conduct daily calibrations with gas standards, but as mentioned in Mynard et al., 2025, the autocalibration system failed while underway, meaning that there were only nine usable calibrations that were conducted manually (every 5 days of the voyage). At the same time, we were conducting mesocosm experiments, so we needed to optimise ambient/mesocosm sampling periods versus downtime periods from calibration/zero measurements. That being said, our calibrations were stable across the voyage and during postvoyage calibrations at KCG. I have added in calibration factor standard deviations for MISO and KCG datasets in the Table S3 to reflect this.

Referee Response 2: Can the authors please add to table S3 that the error stated in the table reflects the standard deviation. The standard deviation of the sensitivity is about 20 % of the mean, so the range is probably larger. This needs to be propagated into the estimate of measurement uncertainty. The authors have increased the estimate of measurement uncertainty from 5 % to 11-12 % during revisions. If the authors could just justify their choice as to how the scatter in calibration curves influences their estimates of uncertainty, that would be appreciated.

Author Response 2: Thank you for these suggestions. We have added to the caption of Table S3 "Values are mean ± 1 standard deviation". Please note for the MISO campaign, the relative standard deviation from the mean is 8 %, as opposed to approximately 20 %. We believe the reviewer has mistaken the 5% uncertainty in the VOC concentrations contained in the calibration gas cylinder (as was first reported coincidentally in Table S3 in another publication – Mynard et al. 2025) as opposed to the 11-12% propagated uncertainty in the ambient VMS measurements; which is unchanged from the first submission of this manuscript/SI. Within this propagated uncertainty, we do account for this calibration variability according to Dunne et al., 2018's PTR-MS uncertainty calculations. We have updated the Supplementary Information with this text:

"Within this propagated uncertainty, we account for uncertainties associated with calibration and ambient measurements, including gas standard concentrations (5%), gas standard flow via mass flow controllers (2%), dilution gas flow via mass flow controllers (2%), ambient ion signal (6%), calibration gas ion signal (2%), and calibration factor variability (relative standard deviation of 8%). These independent uncertainty components were propagated in quadrature, resulting in an estimated combined relative measurement uncertainty of 11-12% for all VMS species. DMS calibrations conducted over multiple calibration gas concentrations during the voyage were consistently linear ($R^2 = 0.9988 \pm 0.0007$; slope = 518 ± 43 cps $ppbV^{-1}$). The between-calibration variability in calibration factors was included as an independent uncertainty term because ambient concentrations were calculated using the temporally nearest calibration factor (Fig. S19)."

See the included Figure S19 in the SI.

Dunne, E., Galbally, I. E., Cheng, M., Selleck, P., Molloy, S. B., & Lawson, S. J. (2018). Comparison of VOC measurements made by PTR-MS, adsorbent tubes-GC-FID-MS and DNPH derivatization-HPLC during the Sydney Particle Study, 2012: A contribution to the assessment of uncertainty in routine atmospheric VOC measurements. Atmospheric Measurement Techniques, 11(1), 141–159. https://doi.org/10.5194/amt-11-141-2018

Supplement

Referee Response 1 L4: How frequent were the backgrounds?

Author Response 1: Zero measurements were conducted during the same time period as the VOC calibrations, approximately every 5 days during the voyage from measurements. Updated to "Ambient data were background corrected by linear interpolation of all nine background measurements to match ambient data cycles and then subtracted to give background-corrected data for each analysed VOC."

Referee response: Some readers might think that this is a very low frequency of zero air measurements – every 5 days. Again, could the authors include some information to show that (1) the background was relatively stable during the campaign and (2) the mean signal to noise ratio was very high meaning the background did not influence the reported concentrations?

Author Response 2: Thank you for these suggestions. We have added a new Supplementary Figure (Figure S19) showing the DMS and MeSH zero measurements throughout the campaign. The updated text in the SI now states from L7: "Figure S19 demonstrates the stability of the instrument response throughout the voyage, illustrating examples of the zero signals and calibration factors applied to the DMS and MeSH measurements. The zero measurements of DMS and MeSH remained stable throughout the campaign, with mean signals of 5 ± 2 cps and 1 ± 0.5 cps, respectively. These background counts per second signals were negligible relative to ambient signals and were therefore unlikely to contribute substantially to the reported background-corrected VOC volume mixing ratio concentrations (Fig. S19)."

Please note that no adjustments have been made to the main text manuscript in this round of responses, only these additions to the Supplementary Information.