We thank the reviewers for their careful reading of the manuscript and for their thoughtful suggestions for improvements. In the following pages, we have given the reviewer comments in regular text, our response in italics, and changes to the text in red.

**Responses to Reviewer 1:**

Line 25: Suggest revise to " in a global atmospheric chemistry-climate model applications/assessments," as it has not actually been included in a climate model

*We agree. The revised text now reads:*

> "Inclusion of methanethiol (MeSH) in a global atmospheric chemistry–climate model assessment has shown that MeSH emissions increase $SO_2$, particulate sulfate and aerosol cooling over the Southern Ocean."

Lines 49-52: Try splitting sentence in 2.

*We agree. The revised text now reads:*

> "This could be due to an under representation in the models of natural marine aerosol forming processes (McCoy et al., 2015), such as through sulfuric acid and methane sulfonic acid production from the oxidation of ocean-derived dimethyl sulfide (DMS, Hossain et al., 2024). Alternatively, it could be due to missing formation pathways involving other species, such as organics, iodine or amines (He et al., 2025)."

Line 54: Suggest changing "chemistry-climate modelling" to "atmospheric chemistry – climate modelling"

*We agree. The revised text now reads:*

> "and subsequent atmospheric chemistry–climate modelling suggests that the inclusion of MeSH emissions reduces a small proportion of this radiative bias (Wohl et al., 2024)."

Line 88: Possibly refer to Ishino et al.

*We agree. The revised text now reads:*

> "could lead to new particle formation and particle growth (Ishino et al., 2026)."

Line 97: Suggest changing "predictions" to "projections"

*We agree. The revised text now reads:*

> "and more informed projections of how their emissions will evolve"

Line 103: Strikethrough "the"

*Revised text now reads:*

> "from 14 February to 17 March 2023"

Line 109: Suggest changing "partway" to "part way"

*"Partway" is standard American English usage but we can change it. The revised text now reads:*

> "The ship then carried out a transect part way towards"

Line 129: Explain Fv:Fm in figure caption and in text.

*We agree. The Figure 2 caption now reads:*

> "(d) photosynthetic active radiation (PAR) and Fv:Fm, a measure of photosynthetic efficiency."

*The methodology for measuring Fv:Fm is described in lines 136 -138 and we have added an additional sentence to explain the meaning of the measurement:*

> "Fv:Fm varies based on phytoplankton population composition in an unpredictable way, but for a given population composition, a low Fv:Fm ratio is thought to indicate phytoplanktonic light stress as an average across the population (Tan et al., 2019)."

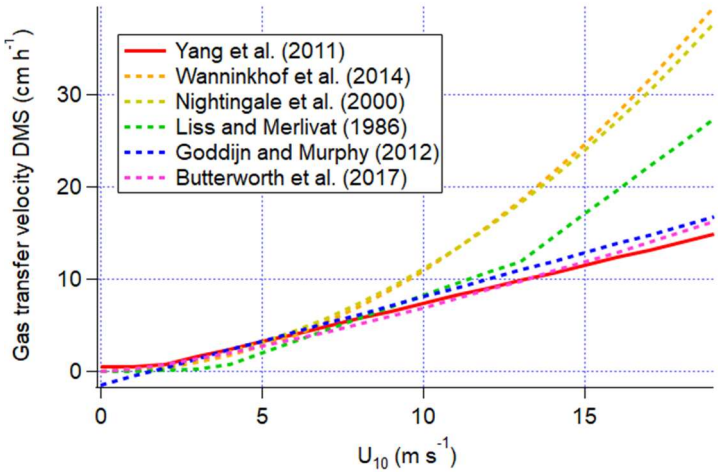
Line 185: Clarify this is atmospheric

*The changes in water reflect changes in atmospheric RH but the changes in pressure are due to the specific setting of the pumping valve and unrelated to atmospheric pressure. We have revised the description of the calibrations to make this clearer. The revised text now reads:*

> "(AIM) reactor held at a fixed pressure between 85 and 95 mbar. The inlet was zeroed every 4 hours with dry gas from a nitrogen generator (LNI). Calibration for DMS was performed after the campaign using the same calibration gas as for the Vocus PTR. The calibrations were conducted over a range of relative humidities (RH) and used to correct the data for changes in sensitivity due to changes in water vapor concentration in the AIM reactor. In particular, the difference in sensitivity between the dry zeros and the ambient measurements (RH 46 to 100 %) needed to be accounted for in the calibrations. The sensitivity also depends on the pressure in the AIM reactor and calibrations were performed at the three pressures used during the campaign to correct for this."

Line 205: A figure comparing this with often used functional k relationships, e.g., Nightingale, Waninkof woud be informative.

*A discussion of the impact of the choice of gas transfer velocity parameterisation on the reported fluxes is beyond the scope of this manuscript. However, in response to the reviewer's comment, we show a figure comparing different gas transfer velocities for DMS. These values were calculated using a fixed Schmidt number of 3466.22, corresponding to the cruise-mean value.*




*The figure demonstrates that gas transfer velocities broadly fall into two categories: exponential and linear parameterisations, with the largest differences occurring at higher wind speeds. The linear parameterisations plotted here (Yang et al. (2011), Goddijn and Murphy (2012) and Butterworth et al. (2017)) are broadly similar and are derived specifically from DMS eddy covariance measurements. As DMS is more soluble than the tracer gases used to derive exponential parameterisations ($CO_2$, He/$SF_6$), its behaviour differs under increasing wind conditions. In particular, at higher wind speeds, DMS gas transfer does not exhibit the same enhancement due to bubbles, resulting in an approximately linear relationship between DMS gas transfer velocity and $U_{10}$. See Blomquist et al., JGR Oceans, 122, 8034–8062, 2017 (https://agupubs.onlinelibrary.wiley.com/doi/full/10.1002/2017JC013181) for more details.*

Line 279: Insert a comma before “too”

*We have added the comma.*

Line 301: Why switching between ppbv in figure and pptv in text?

*We agree that the units should be consistent between the figure and the text and have updated all numbers in pptv to ppbv. We have also updated Table S1 to ppbv. The revised text reads:*

> “The mean MeSH ambient air concentration was 0.015 ppbv with a range of 0.001 to 0.060 ppbv. These MeSH air mixing ratios are similar to previous measurements in marine air of the North East Pacific (0.019 ppbv, Novak et al., 2022), the eastern North Atlantic (0.015 ppbv, Kilgour et al., 2024), the North Sea (0.016 ppbv, Wohl et al., 2025), the southwest Pacific Ocean (0.018 ppbv, Lawson et al., 2020) and the Southwest Pacific Ocean east of New Zealand (0.040 ppbv, Rocco et al., 2025). Our measurements are within the range of MeSH observations over the Southern Ocean near the sea ice edge binned by latitude between 62° and 66° S (medians 0.014–0.064 ppbv, Mynard et al., 2025).”

Line 310: Why ppb not ppbv as in previous figure?

*The axis labels in Figure 7 have been corrected.*

Line 334: Correct bracketing.

*This was a quirk of the reference manager (EndNote) which can’t put superscripts inside the parentheses. We have revised the text to read:*

> "in coastal temperate waters of 0.74 µmol $m^2$ $d^{-1}$ (Novak et al., 2022), estimates from the nighttime accumulation technique of 0.36 – 4.14 µmol $m^2$ $d^{-1}$ (Rocco et al., 2025), and two-layer fluxes calculated using air and seawater measurements in the Great Barrier Reef of 1.2 µmol $m^2$ $d^{-1}$ (Deschaseaux et al., 2025)."

*We have revised lines 229-233 which had the same issue. The revised text reads:*

> "Despite the large uncertainty, this average concentration was higher than average measurements across the Atlantic of 0.39 nmol $dm^{-3}$ (Kettle et al., 2001), near the Mediterranean coast of 0.3 nmol $dm^{-3}$ (Wohl and Simó, 2024) and in the Great Barrier Reef of mean 0.49 in 2021 and 0.34 nmol $dm^{-3}$ in 2022 (Deschaseaux et al., 2025). These MeSH concentrations were lower than measurements in the Arctic of 2.96 nmol $dm^{-3}$ for polar water (Gros et al., 2023) and similar to measurements in the sub-Arctic of 0.57 nmol $dm^{-3}$, average of cruise OC1607 and OC1708 (Kiene et al., 2021), where episodes of higher than 1.5 nmol $dm^{-3}$ MeSH were also observed."

Line 343: Something missing in this sentence.

*We have revised the sentence to read:*

> "For MeSH, seawater concentrations were on average 60 times larger than $C_aH$, indicating that the seawater was strongly supersaturated relative to the atmosphere. As a result, the thermodynamic driving force consistently favoured sea to air transfer."

Line 353: Not visually obvious. As significance has been tested, I suggest to mention somehow that the diel cycle is visually not very obvious, but still significant, especially since the DMS plots don't appear too different, but are not significant.

*We have revised the text to make it clearer that, although the plots look similar, diel change in MeSH is statistically significant while the diel change in DMS is not.*

> "Although not visually pronounced in the cruise-mean data, we observed statistically significant diel variability in surface seawater concentrations, sea–air fluxes, and ambient air mixing ratios of MeSH (Fig. 9)."

> "In contrast to MeSH, the surface seawater DMS concentrations over the entire cruise did not show a statistically significant diel change, although"

Line 422: Note what this means with respect to significance and correlation.

*The revised text reads:*

> Values in bold show that the two-tailed p value of the correlation is less than 0.01, indicating statistically significant linear correlation.

Line 457: Would that mean that the relevance of MeSH would be less than suggested by Wohl et al., 2024?

*We are unable to comment on this without further extensive model runs, which are unfortunately beyond the scope of this manuscript.*

Line 466: Also limits application of relationship-based climatologies (maybe suggesting that dynamical models may be helpful).

*We agree that dynamical models, independent of DMS climatology, would be helpful for calculating MeSH seawater concentrations. However, it is not clear that we can draw these conclusions from our limited observational dataset.*

**Responses to reviewer 2**

Line 149: Are potential contamination from the tubing supplying tap water possible? Additionally, were any measures taken to prevent biofouling in the tubing? Please clarify these points in the manuscript.

*To check for biofouling or contamination in the ship's underway water supply, we compared DMS and MeSH concentrations measured from the ship's underway water supply to those simultaneously measured from the CTD Niskin at 5 m as discussed in Section 4 of the Supplemental. This uses the CTD Niskin at 5 m as a reference indicating sampling without contamination. We have revised the text to make this clearer:*

> "Seawater MeSH concentrations are not reported for the first part of the cruise due to bacterial growth in the SFCE causing unrealistically high concentrations (see Sect. S3). This issue was dealt with by daily rinsing of the tubing with 3.7% HCl. Thereafter, we checked for bacterial growth in the SFCE and the underway seawater supply lines of the ship by comparisons to concentrations measured in the 5 m Niskin bottle from the CTD casts. Both MeSH and DMS agreed well between the underway seawater supply and the 5 m Niskin bottle indicating that biofouling and contamination were minimal (see Sect. S4)."

Line 158. Could the daily cleaning procedure affect phytoplankton within the tubing and influence the measured concentrations? Additionally, were blank water measurements performed? If so, could the authors provide these data?

*The cleaning procedure is discussed in more detail in Wohl et al. (2019). The relevant section is copied here for convenience:*

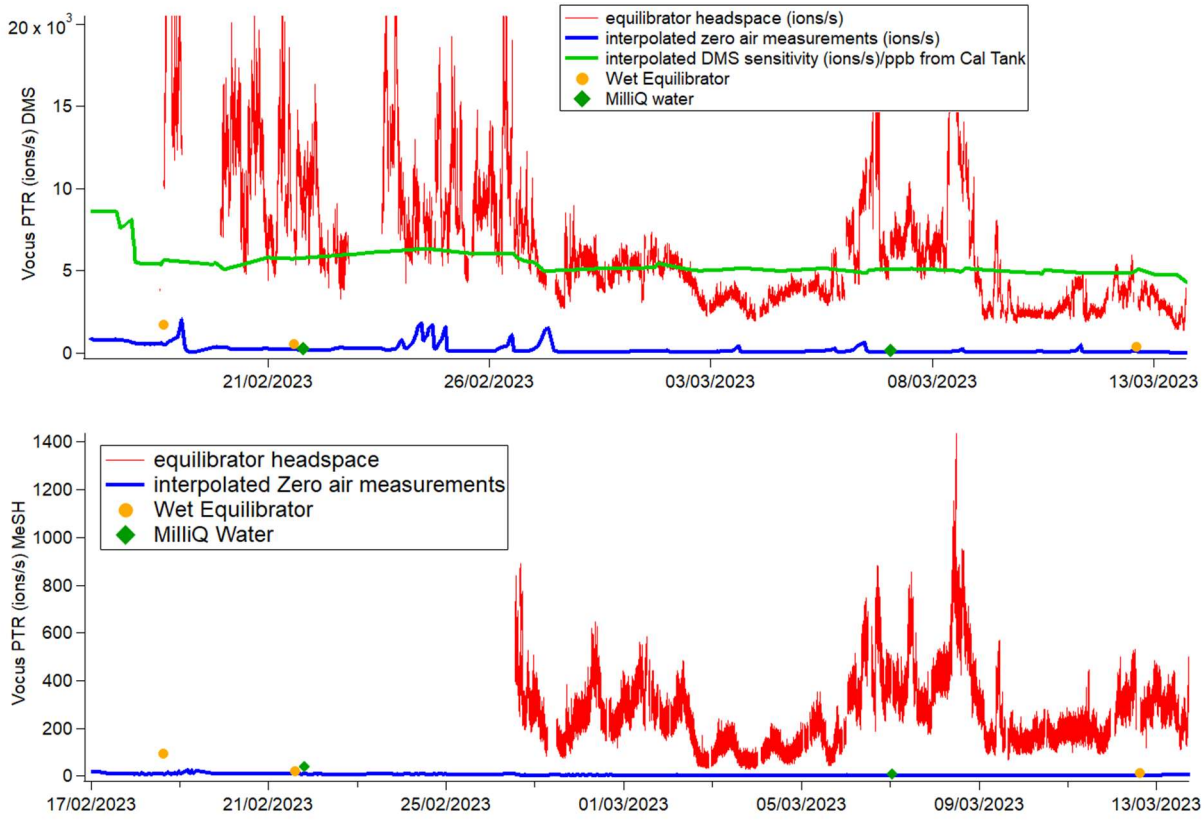
*"Due to the smooth surfaces and constant and complete water renewal, the equilibrator should not be very prone to biofouling. The lack of a membrane for gas exchange means that the degree of equilibration should not vary significantly even if there is some biofouling. To clean the SFCE if necessary, the seawater intake and the water drain pipe are connected to a 10 % HCl solution for 10 min. During this procedure, the PTR-MS is disconnected to sample lab air and the gas flow is stopped. A flow of HCl thus covers all the parts of the equilibrator that are normally exposed to seawater. To resume measurement of ambient seawater, the flow of HCl is stopped and the carrier gas flow is started to drain the HCl safely into the recirculated solution. The equilibrator is typically rinsed with seawater before resuming measurement."*

*Because the SFCE is rinsed with seawater after HCl cleaning, the conditions experienced by phytoplankton before and after cleaning should be equivalent. In addition, the seawater residence time within the SFCE is short (approximately 30 seconds under the flow conditions used here). Given this brief residence time, any impact of the cleaning procedure on phytoplankton is expected to be minimal.*

*Regarding background measurements, the only blanks collected regularly were hourly zero-air measurements. These were obtained by measuring air from a zero-air generator (Vocus PTR Clean Air System, Tofwerk AG) directly with the Vocus PTR.*

*In earlier deployments using an older Ionicon quadrupole PTR-MS, a wide range of blank types was collected, and reported concentrations were sensitive to the choice of blank. However, laboratory tests prior to this deployment showed that many of these blanks produced very similar results with the newer Vocus PTR. The Vocus PTR is insensitive to humidity effects and, due to its time-of-flight (ToF) capability, can resolve many interferences that previously required correction using water-based blanks.*

*To maximise data collection during the campaign, blank measurements were therefore limited to periods when scheduling allowed. The figures below compare wet equilibrator blanks and Milli-Q water blanks for DMS and MeSH with interpolated hourly zero-air measurements and seawater equilibrator headspace measurements. All signals are shown in raw instrument counts (ions $s^{-1}$) to enable direct comparison of signal and background.*




*These figures demonstrate a high signal-to-background ratio and show that SFCE-specific water blanks (MilliQ and wet equilibrator) closely match the more frequently acquired zero-air measurements. Consequently, the calculated concentrations are not strongly dependent on the choice of blank. Given their higher temporal resolution, hourly zero-air measurements were used as the blank for both DMS and MeSH.*

*We have revised the description of the background subtraction in Section S2 to include a few more details. The revised text reads:*

> "Hourly measurements of zero air were used for background subtraction. We note that for DMS and methanethiol, we observe good agreement between the zero air measurement and a wet equilibrator blank. The wet equilibrator blank consists of stopping the water flow into the equilibrator and flushing it for 1 hour with zero air. We also performed occasional blanks by measuring MilliQ water with the SFCE using the same methodology as for discrete samples. These blanks also agreed well with the zero air measurements. Therefore, we used the more frequent zero air measurements for background subtraction. We note that DMS and methanethiol have a high signal to background ratio; thus, reported concentrations are relatively insensitive to the choice of background."

Line 172: Please clarify how MeSH was calibrated using the permeation method. Was a permeation tube used in an oven, and if so, at what temperature? Additionally, was the concentration generated from the permeation tube diluted prior to measurement?

*We have added a few more details about the MeSH calibration. The revised text reads:*

> Calibration for MeSH using a permeation tube was performed after the campaign (Vici, emission rate of 103.8 ng/min at 40 C, held in a perm tube oven at 40 C, diluted with 2 to 5 slpm of zero air).

Line 220: 1.6 nmol dm-3 +- … ? Please give the uncertainty (standard deviation?)

Line 226: As for MeSH average concentration, give the uncertainty. Please check for all other values where uncertainty can be added (l.388 to 389 for example).

*Giving the range of the measurements, rather than the standard deviation, seemed more useful for describing atmospheric conditions. The uncertainty and the standard deviation for a time-varying quantity are not the same thing. The measurement uncertainties for the seawater measurements are discussed in Section S2 and for the air measurements are discussed in Section S5. The standard deviations of the time series measurements are given in Table S1. The text has been revised to make this clearer:*

> *Lines 239-240 of revised manuscript:* "The estimated uncertainty for DMS is ± 20% (see Sect. S2). Additional statistics, such as median, standard deviation, 25$^{th}$ percentile and 75$^{th}$ percentile, are given in Table S1 for the seawater and air measurements presented here."

Line 257. Change ")(" as ";"

*Typo is fixed.*

Line 298. What are these values: "(0.011 nmol dm-3)" , "(0.0004 to 0.059 nmol dm-3)? Uncertainty?

*Since air measurements are typically reported in ppbv and seawater measurements are typically reported in nm dm$^{-3}$, we thought it would be useful to report the air measurements in the same units as the seawater measurements. This is shown with the right axis units in Fig. 6 and in parentheses. However, if this is confusing, we can remove the nm dm$^{-3}$ values from the text. The revised text reads:*

"Mean DMS ambient air concentration measured with the Vocus PTR was 0.25 ppbv with a range of 0.008 to 1.35 ppbv."

"The mean MeSH ambient air concentration was 0.015 ppbv with a range of 0.001 to 0.060 ppbv."

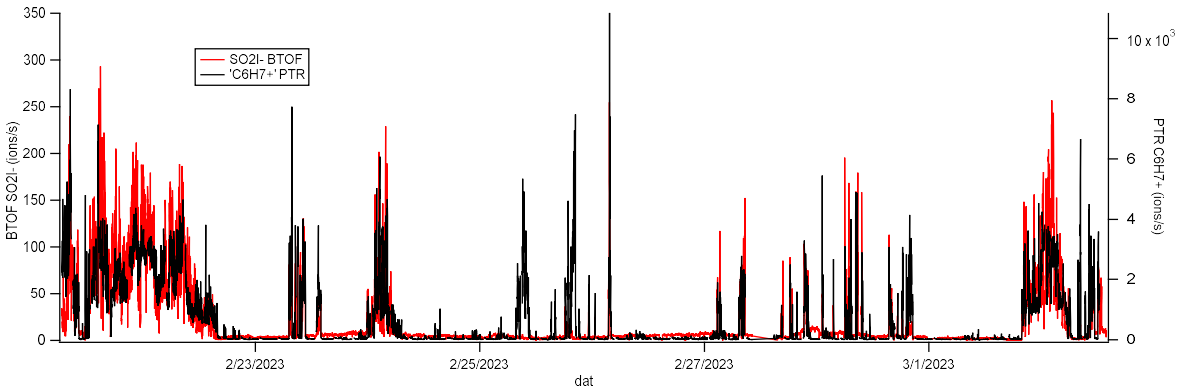
Supplementary material:

Line 110. It is stated that cleaning was performed with 3.7 % HCl. In Wohl et al. (2024), it was stated that was 10%. Please clarify.

*Hydrochloric acid (HCl) was purchased at a concentration of 37% and diluted tenfold before use. Although this solution was historically labelled as "10% HCl" in the laboratory, the actual concentration of HCl after dilution is 3.7%. This labelling error has now been corrected. The lower, correct concentration of HCl used for cleaning is not expected to significantly affect our measurements.*

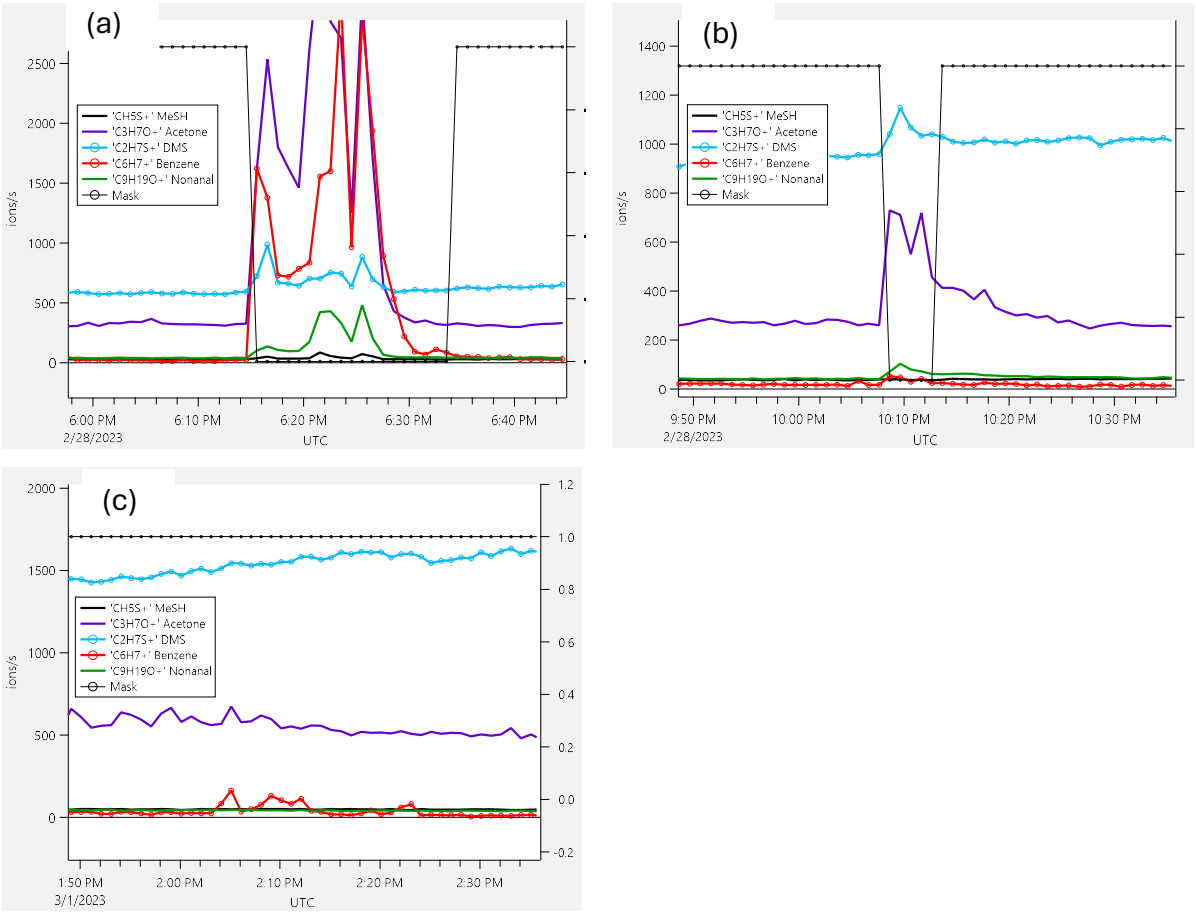
Line 169. Can you check the potential pollution by ship exhaust with SO2 measurements?

*Yes, we can see evidence of ship exhaust in the $SO2I^-$ signal from the $I^-$ channel of the BTOF. The figure below shows a time series comparing the benzene signal ($C6H7^+$) from the PTR with the $SO2I^-$ signal from the BTOF. The benzene signal identifies a few more periods of ship exhaust than the $SO2I^-$ signal. Therefore, we used the benzene signal to remove time periods contaminated with ship exhaust.*



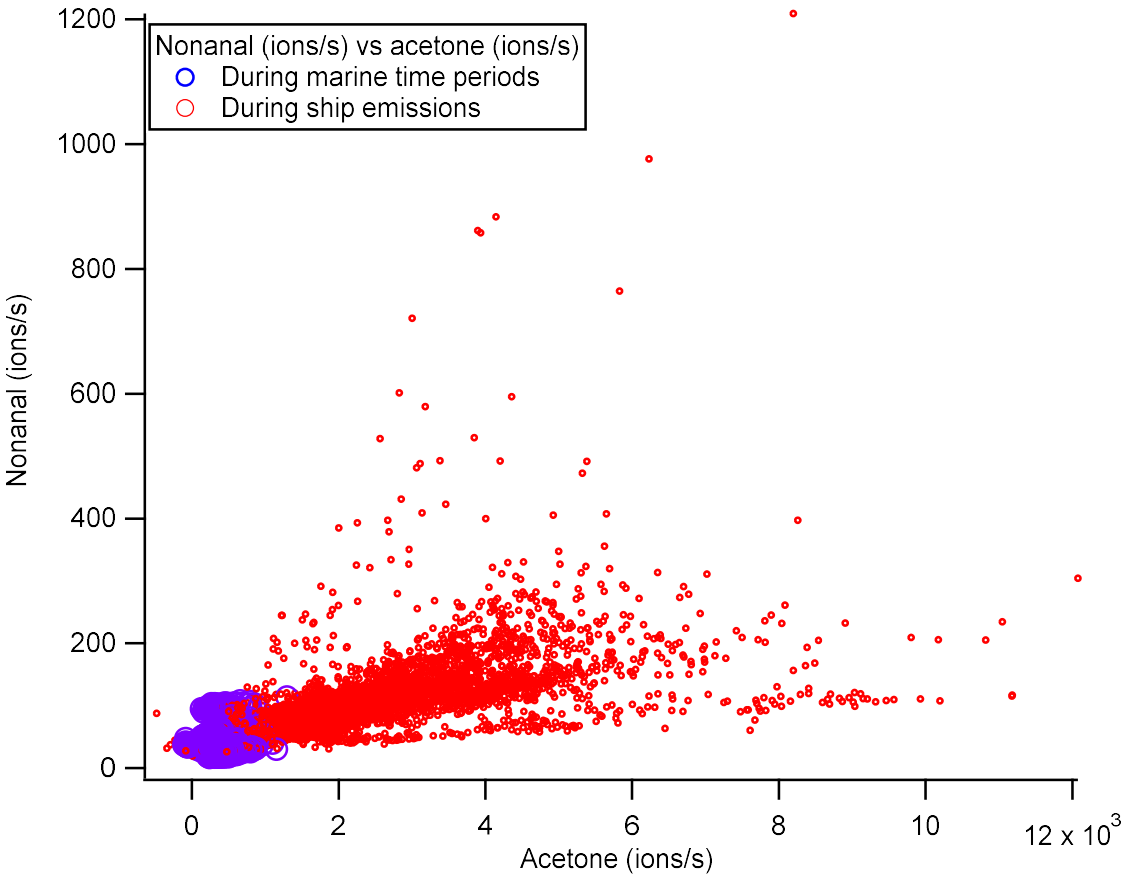

Line 171. Do the authors consider that nonanal and acetone may have a biological origin that could explain their interference with DMS and MeSH concentrations? Additionally, what are the correlations between these compounds during the excluded periods compared to the retained dataset?

*The nonanal and acetone signals were sharp and spiky during the time periods that were contaminated with ship emissions (based on the presence of benzene). During periods of sampling marine air, nonanal and acetone were lower and varied slowly. Panel (a) in the figure below shows a typical time period likely contaminated with both kitchen and engine exhaust*




*based on the presence of benzene and nonanal. The DMS and MeSH signals show spikes that follow the acetone/benzene/nonanal spikes and are due to interfering peaks at m/z 63 and 49 that grow in and bias the peak fits high. Most of the time periods removed for ship emissions looked like this. There were only a few time periods eliminated on the basis of only acetone and nonanal. An example is shown in panel (b). In addition to the spikes in acetone/nonanal which are not characteristic of marine time periods, there is a hint of benzene indicating that this is ship emissions rather than biological activity. For completeness, panel (c) shows a time period when there was some benzene present but there were not spikes in nonanal or acetone. These time periods did not show interference in DMS or MeSH and were retained in the time series.*

*The next figure shows a correlation plot of nonanal and acetone with the marine air points in blue and the ship emissions points in red. The slope looks similar but the absolute magnitudes of nonanal and acetone are higher during ship emissions.*




**Minor edits requested by co-author**

*We made some minor edits to Section 2.2 on the biological measurements. These edits were received from a co-author after the manuscript had been submitted. The revised text reads as follows:*

> Biological and biogeochemical measurements were conducted every six hours over most of the cruise at 06:00, 12:00, 18:00 and 24:00 local time. A brief overview of the methodology is presented here while extensive details are given in Rocchi et al. (2025). Seawater samples were collected from the underway water inlet for the estimation of bacterial (2.0 $cm^3$, preserved with 1 % paraformaldehyde plus 0.05 % glutaraldehyde, final concentration) and pico- and nano-phytoplankton (5.0 $cm^3$, preserved with glutaraldehyde, 1 % final concentration) cell abundances. All samples were stored at -80 C until analysis at the ICM-CSIC within 5 months. A Cytoflex flow cytometer was used for bacterial number estimation and a CyFlow Cube 8 (Sysmex) was used for pico- and nano-plankton abundances.