Response to Reviewers for: "Oceanic emissions of dimethyl sulfide and methanethiol and their contribution to sulfur dioxide production in the marine atmosphere"

We thank both reviewers for the constructive comments on our manuscript. Reviewer comments are reproduced below along with author responses and any significant changes made to the manuscript text.

Reviewer comments are in green

Author responses are in black

Additions to the text are marked in red, and deletions are shown with a red ~~strikethrough~~

Reviewer 2:

The paper entitled "Oceanic emissions of dimethyl sulfide and methanethiol and their contribution to sulfur dioxide production in the marine atmosphere" is novel, interesting and falls within the scope of ACP. The authors report the first direct eddy covariance flux measurements of MeSH oceanic emissions and perform a comprehensive analysis of the implications of this findings with the help of a chemical model.

In general, the paper is well written and I have only a few minor comments:

We thank the reviewer for their review and suggestions which have improved the quality of our paper!

1. Section 1.1 It would be nice if the authors include the reference to some more recent studies elucidating the methanethiol production pathways by Sun et al. (2016)

   Agreed, discussion of the Sun et al. work has been added.

   Line 63: "The bacteria *Pelagibacter* HTCC1062 has been shown to simultaneously produce both DMS and MeSH, where the allocation between products may be related to the available supply of DMSP, with DMS production enhanced when the supply of DMSP exceeded the cellular demand for sulfur (Sun et al., 2016)."

2. The work of Sun et al. (2016) should also be discuss the study in the discussion line 353ff.

   Yes, discussion of the Sun et al. (2016) work is warranted here and has been added.

   Line 364: "Sun et al., (2016) have also shown that the bacterium *Pelagibacter* produces both DMS and MeSH from DMSP, where the relative yield of products is related to the amount of excess DMSP compared to the cellular demand for sulfur for biosynthesis."

3. Section 1.3 Can the reaction mechanism be displayed in some form of graphic/schematic? This is a little hard to follow

A reaction diagram has been added as Figure 1.

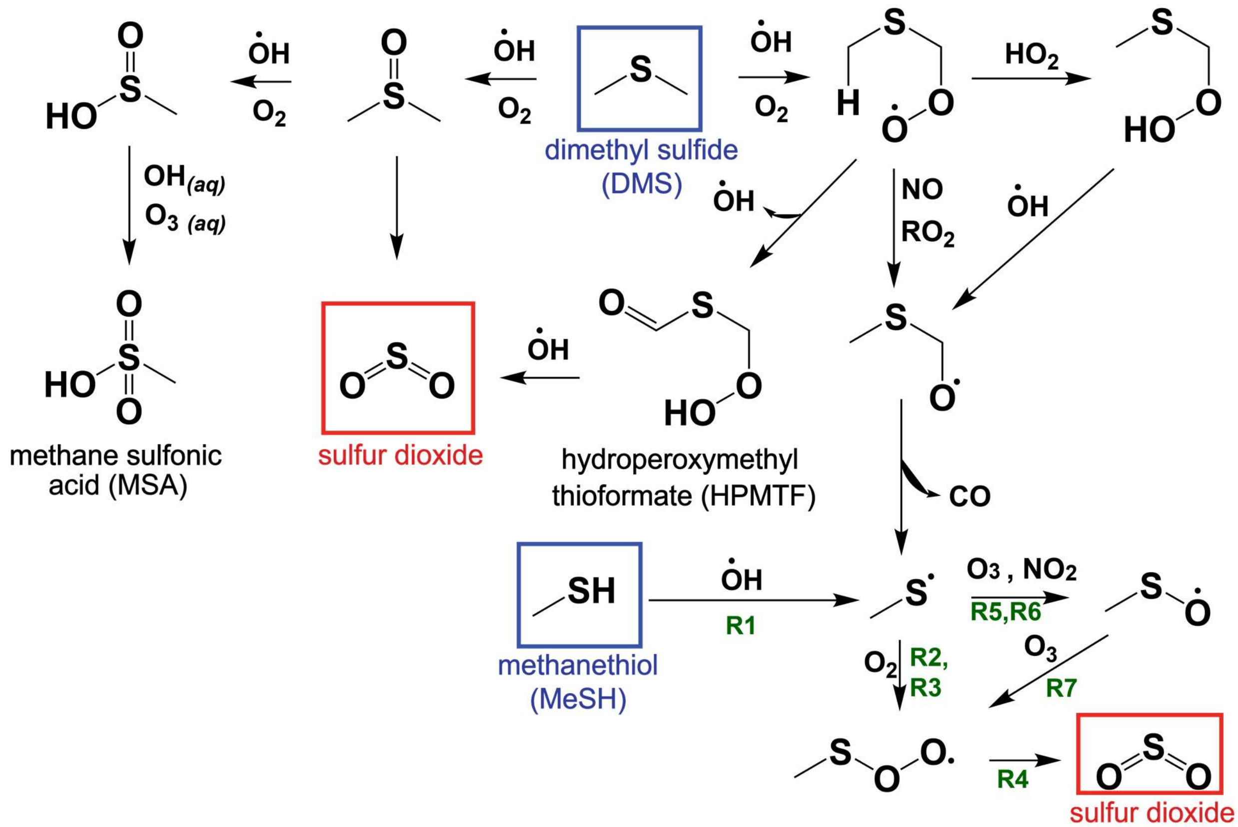

Figure 1. A simplified reaction scheme for the gas phase oxidation of dimethyl sulfide (DMS) and methanethiol (MeSH) that focuses on pathways to SO_2 production. Reactions R1 through R7 described in Section 1.3 are labelled with green text on the schematic. Other chemical pathways including oxidation by halogens and most condensed phase reactions of DMS and its oxidation products are not shown in this simplified schematic. Refer to Table S1. for a complete list of reactions and rate equations as implemented in this work.

4. Methods section: I seem to be unable to find the section where the meteorological measurements and the equipment used for the eddy covariance flux system are described. Can the authors add this description?

   A description of the meteorological measurements for the eddy covariance system were given at line 77 “The ambient inlet sampling point was collocated with a sonic anemometer recording three-dimensional winds at 10 Hz (Gil HS-50). The sonic anemometer and Vocus inlet were mounted on a 6.1 m long boom extended beyond the end of the pier to minimize flow distortions from the pier. The inlet was mounted on the boom at a height of 13 m above the mean lower low tide level.”

5. Line 449 the recent ship cruise in the Arabian Sea was not the first study dimethyl sulfone DMSO2 has also been reported in marine air masses in Antarctica. Berresheim et al. 1998 reported it **https://doi.org/10.1029/97JD00695**

Thank you for bringing this to our attention, we have added some discussion of the Berresheim et al. observations.

Line 468: "$DMSO_2$ has also been measured at Palmer Station, Antarctica in January to February of 1994 with mean and median mixing ratios of 1.7 and 1.3 ppt respectively (Berresheim, 1998). The higher $DMSO_2$ mixing ratios observed in that study are likely at least in part due to the much lower temperatures (mean 274.5 K), where the DMS + OH addition channel forming DMSO and $DMSO_2$ is more favored."

Additional Comment:

Subsequent to submitting this manuscript, a paper was published providing the first measurement of the HPMTF + OH rate constant ($k_{HPMTF+OH}$). $k_{HPMTF+OH}$ was found to be 1.4 (0.27 – 2.4 uncertainty range) × 10^{-11} cm^3 $molec^{-1}$ s^{-1}, which is consistent with the rate constant of 1.1 × 10^{-11} cm^3 $molec^{-1}$ s^{-1} used in this work estimated from the structurally similar molecule methyl thioformate. We add some discussion of this new measurement in SI section S5 where we discuss HPMTF chemistry.

SI Line 104: "The bimolecular rate constant of HPMTF with OH ($k_{OH+HPMTF}$) was approximated to be 1.1 × 10^{-11} × 10^{-11} molecules cm^{-3} s^{-1} which is the rate of OH + methyl thioformate which is structurally similar molecule to HPMTF, which is within the uncertainty range of a recent laboratory determination of 1.4 (0.27 – 2.4 uncertainty range) x 10^{-11} cm^3 $molec^{-1}$ s^{-1} (Jernigan et al., 2022) ~~as $k_{OH+HPMTF}$ has not been experimentally determined~~."