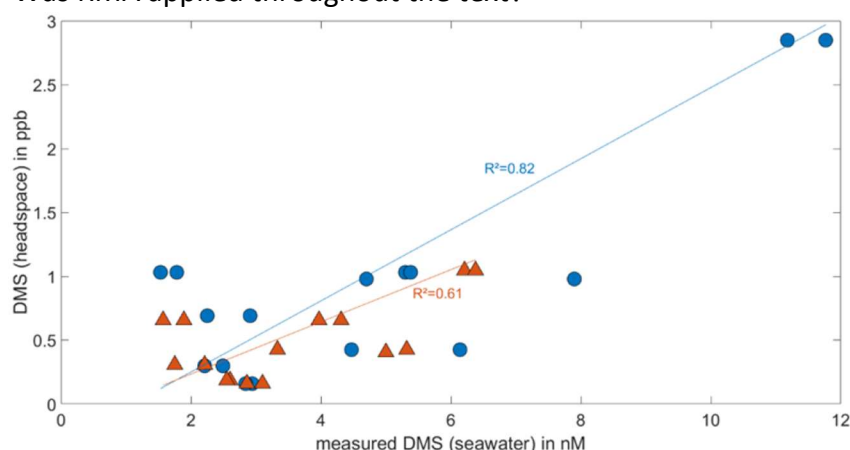


Neither in its form nor in its scientific content is this manuscript suitable for publication in Atmospheric Chemistry and Physics (ACP).

**In its form.** My overall feeling is that this manuscript has been written in a rush and, contrary to the author contributions statement, all authors very likely did not contribute to its writing, review and editing, e.g.:

- 2 different titles: in the main text the MS is entitled “Air-Sea fluxes of dimethyl sulphide and methanethiol in the South-West Pacific” whereas in the supplementary it is entitled “Sea-Air fluxes of dimethyl sulphide and methanethiol from mesocosm studies of natural seawaters from the South-West Pacific”.
- Reference list: About 17 references cited in the reference list are not cited in the main text. About 12 references cited in the main text are not listed in the reference list, including the recent contributions of the corresponding author (Sellegri et al., 2022; Sellegri et al., 2023)!
- Data displayed in Fig. 4 and Fig. 5 are inconsistent. DMS mixing ratios are 10 times lower in Fig. 4 than in Fig. 5.
- Fig. S.1: Although RMA regression instead of least-squares regression has been used in Fig. S.1 (shown below), I doubt that the authors can conclude that “the correlations between DMS in air and water did not differ between the ASIT-control and ASIT-O3”. The positive blue regression (applied to ASIT-control isn’t it?) is in fact driven by only two data points. In terms of slope, intercept and r2 values, why are different numbers provided in text and Fig. S.1? Why are the r and r2 values the same? Was RMA applied throughout the text?



ASIT-control : slope =  $0.28 \pm 0.41$ ; intercept =  $-0.31 \pm 0.08$ ;  $r = 0.82$

ASIT-O3: slope =  $0.21 \pm 0.32$ ; intercept =  $-0.17 \pm 0.08$ ;  $r = 0.61$

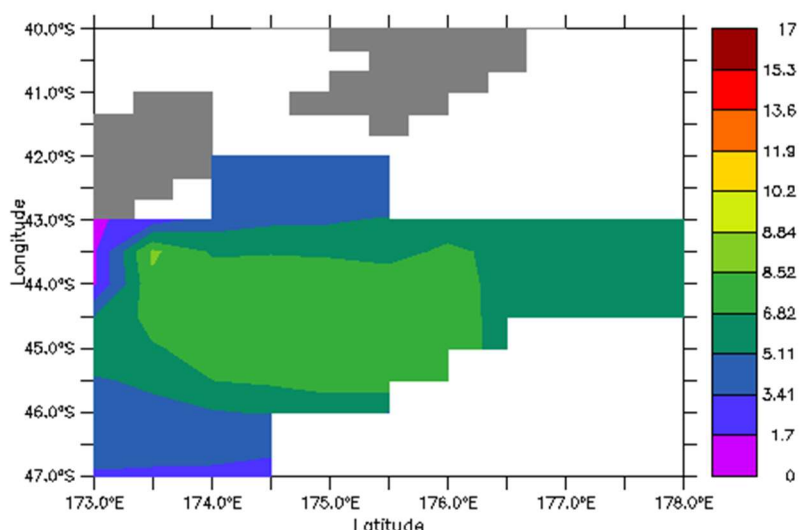
**Figure S. 1: Reduced major axis (RMA) of measured ASITs DMS (seawater) vs. DMS (headspace).**

## In its scientific content.

This work makes insufficient use of DMS state-of-the-art. References to recent studies are missing in the introduction. The Lana et al. (2011) climatology of marine DMS has been updated recently (<https://doi.org/10.5194/essd-14-2963-2022>). It now incorporates more than 1000 data points collected by Bell et al. (2015) in the same area than that investigated during the Sea2Cloud campaign (see below) where only a few tens of samples were analyzed for DMSw in the framework of the ASIT experiments. You should make this clear in the introduction.

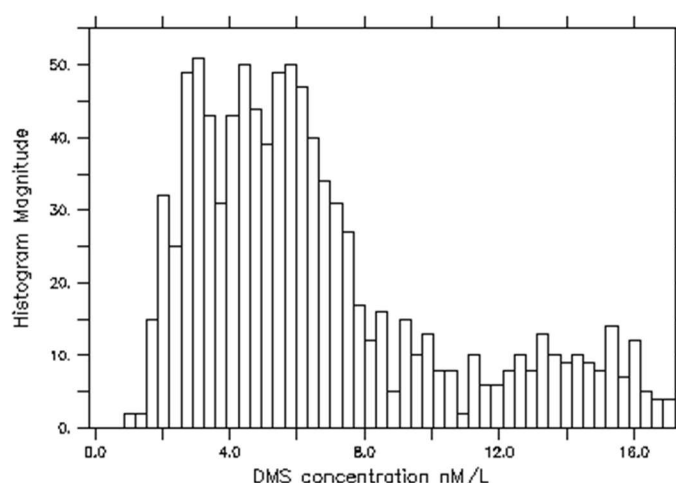
## DMS Database Output

Limits	Minimum	Maximum
Date	1972-03-11	2019-04-15
Latitude	-47	-40
Longitude	173	178
Month(s)	all	



Data Record Statistics	
Number of records	1012
Minimum DMS value	1.10
95th Percentile DMS value	17.05
Maximum DMS value	33.63
Mean DMS Value	7.28
Median DMS Value	5.83
DMS Standard Deviation	4.77

[Download This Data Set](#)  
Please read the [data file description](#)



Cont. Num	Contributor	Platform	Region	Num Samples	Start	End	Reference(s)
247	Bell	R/V Tangaroa	Southern Ocean	1012	2012-02-15	2012-03-06	Bell et al., 2015

In the introduction, it is also stated that “In the surface ocean, DMS is produced from the degradation of dimethylsulfoniopropionate (DMSP), which is produced by marine macroalgae, phytoplankton or bacteria (Bentley and Chasteen, 2004; Kloster et al., 2006; Novak and Bertram, 2020). To form DMS, the DMSP undergoes reactions catalysed by DMSP lyase, (Taylor and Visscher, 1996; Steinke et al., 1996; Kiene, 1996a) and non-enzymatic pathways of demethylation (Bentley and

Chasteen, 2004). Furthermore, DMS can be produced by the biological reduction and oxidation of DMSP and also abiotically by light-dependent reactions (McNabb and Tortell, 2021 and references therein)". According to the very recent review of Shaw et al. (2022, Shaw, D.K., Sekar, J. and Ramalingam, P.V. Recent insights into oceanic dimethylsulfoniopropionate biosynthesis and catabolism. Environmental Microbiology (2022) 24(6), 2669–2700. doi:10.1111/1462-2920.16045), the existence of a "non-enzymatic pathways of demethylation (Bentley and Chasteen, 2004)" is highly unlikely as is "DMS production by the biological reduction and oxidation of DMSP". I acknowledge that the metabolism and catabolism of DMSP is complex as shown below but, unfortunately, the present manuscript does not make any advance in this field. The importance of nanophytoplankton in DMSP synthesis has been established 20 years ago.

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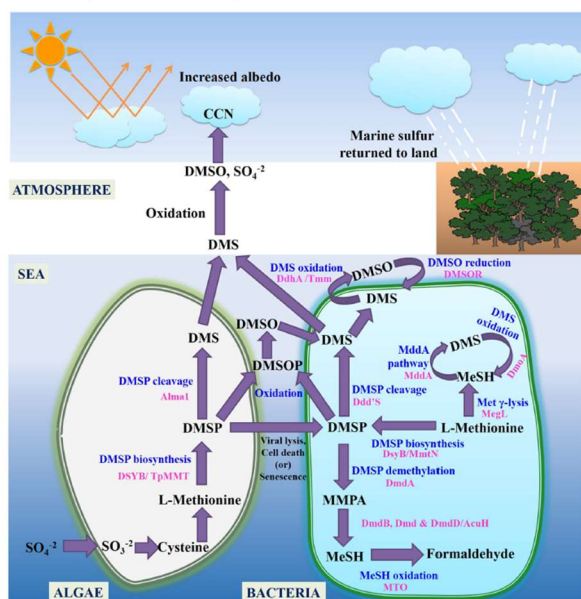


Fig. 4. Microbial biosynthesis and cycling of DMSP and DMS. AcuH, acrylyl-CoA hydratase; AlmA1, DMSP lyase; CCN, cloud condensation nuclei; DMS, dimethyl sulfide; DMSA, dimethylsulfide dehydrogenase; DmdA, DMSP demethylase; DmdB, MMPA-CoA ligase; DmdC, MMPA-CoA dehydrogenase; DmdD, methylthioacrylyl-CoA hydratase; DmdK, dimethylsulfide monooxygenase; DMS, dimethylsulfide; DMSO, dimethyl sulfoxide; DMSOR, dimethyl sulfoxide reductase; DMSY, dimethylsulfoniopropionate; DMSOP, dimethylsulfoxonium propionate; DmsB, DmsY, DmsZ, DmsH, S-methyltransferase; MegL, Methionine  $\gamma$ -lyase; MeSH, methanethiol; MMPA, methylmercaptopyruvate; MTH, Met-methylating enzymes; MTO, MeSH oxidase; Tsm, trimethylamine monooxygenase; TpMMT, methylthiohydroxybutyrate SAM-dependent methyltransferase.

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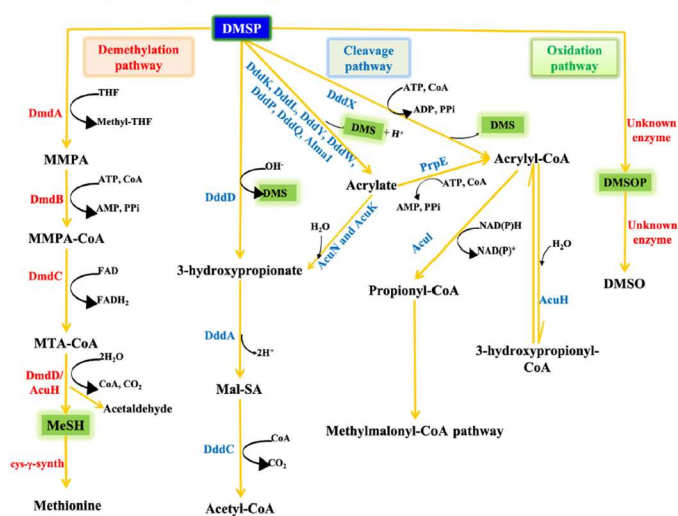


Fig. 3. Biochemical pathways for dimethylsulfoniopropionate catabolism. In the cleavage pathways, several DMSP lyases DddL, DddP, DddQ, DddW, DddK, DddX, DddY or algal AlmA1 catabolizes DMSP to acrylate with the release of dimethyl sulfide (DMS), and acrylate is then converted to 3-hydroxypropionate (3HP) by the action of AcuN and AcuH, whereas the DMSP lyase DddD converts DMSP to 3HP. 3HP is then converted to malonate semialdehyde (Mal-SA) and then acetyl-CoA by DddA and DddC, respectively. An acrylate-CoA ligase (PrpE), an acrylate-CoA reductase (AcuI) and AcuH are also involved in the cleavage pathway. The DMSP demethylation pathway is catalysed by the DMSP demethylase (DmdA), MMPA-CoA ligase (DmdB), MMPA-CoA dehydrogenase (DmdC), and either the MTA-CoA hydratase (DmdD) or acrylate utilization hydratase (AcuH). In the oxidation pathway, DMSP is oxidized to dimethylsulfoxonium propionate (DMSOP). However, enzyme involved in this pathway is unknown.

I agree with Dr. Dong's comment. The sea-air fluxes of DMS and CH<sub>3</sub>SH measured in the ASIT tanks by no means can be compared with literature values reported in Table 3, because the turbulence very likely is considerably lower in the tanks than in the reality. By no means can this manuscript be entitled "Air-Sea fluxes of dimethyl sulphide and methanethiol in the South-West Pacific", and the following sentences "Air-sea fluxes of dimethyl sulphide (DMS) and methanethiol (MeSH) from surface seawater in the remote Southern Pacific Ocean were measured in three Air-Sea Interface Tank (ASIT) experiments during the Sea2Cloud voyage in March 2020. The measured fluxes of  $0.78 \pm 0.44$  ng m<sup>-2</sup> s<sup>-1</sup> and  $0.05 \pm 0.03$  ng m<sup>-2</sup> s<sup>-1</sup> for DMS and MeSH, respectively, varied between experiments reflecting the different water mass types investigated, with lowest fluxes with subtropical water and highest with biologically-active water with sub-Tropical water and highest from the sub-Tropical Front" be put forward in the abstract.

Another major flaw is the role attributed to ozone, i.e. "The experiments also determined the influence of elevated ozone, with one ASIT headspace amended with 10 ppbv ozone while the other provided an unamended control. Elevated ozone resulted in a decrease in DMS flux, corresponding to decreased conversion of dimethylsulfoniopropionate (DMSP) to DMS in the seawater". Concentrations of DMSP and DMS in seawater are displayed in Fig. S.5 and Fig. S.2, respectively (reproduced below). Although the poor quality of both figures makes the comparison difficult to be established, it seems that it is in ASIT-control not in ASIT-O<sub>3</sub> that a major change (an increase in this case) in the DMS-to-DMSP ratio took place in EXP-A by Mar 22. How can one trust that ozone is an inhibitor of the conversion of DMSP to DMS in seawater when the demonstration relies on a single observation? This process should have been investigated in the laboratory under more controlled conditions and in replicates.



Figure S.5: Concentrations of DMSP in seawater (nM).

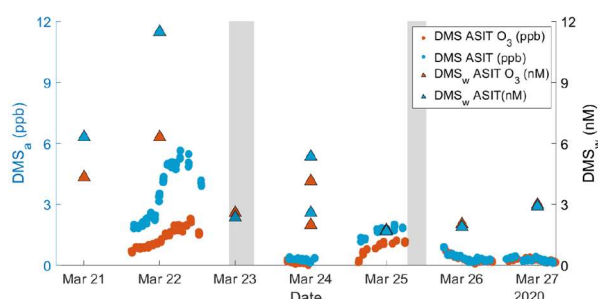


Figure S.2: Concentration of DMS in seawater (nM) and air (ppbv) in ASIT-control (blue) and ASIT-O<sub>3</sub> (orange). ~20min average DMS headspace mixing ratios (ppbv, dots) in ASIT-control (blue) and ASIT-O<sub>3</sub> (orange) and dissolved DMS in ASIT's seawater samples (nM, triangles).

This manuscript should be rejected in its present form.