A point-by-point reply to the comments is given below.

Comments from the reviewer are in black; answer to the reviewer are in blue, new adding to the text are in black and italics.

Review of the paper: "Variability of dimethyl sulphide (DMS), methanethiol and other trace gasesin context of microbial communities from the temperate Atlantic to the Arctic Ocean"

by Valérie Gros et al.

General comments

The authors have undertaken an in-depth revision and have improved considerably most of the article. However, the writing could still be improved throughout, and I recommend a thorough check of English (grammar and orthography) and the References. The work looks solid from a technical standpoint, but I encourage the authors to report whether cross-calibration between underway and Niskin samples was performed. If not, please add a note of caution because Niksin sampling and underway systems can yield somewhat different concentrations, especially in the case of gases (at least DMS) that respond quickly to mechanical stress induced by underway pumping on cells. A final note: this paper contains at least 2 different stories, and the one about DMS and MeSH has been treated more in depth and could make a separate paper. Of course this is the authors' decision.

We thank the reviewer for the positive evaluation of our revised manuscript and hope the paper is now again written in a clear and well-structured way. In this new revised manuscript, we have performed a thorough check of English.

We agree that the paper mostly focus on DMS and MeSH with only little discussion on the other compounds. Nevertheless, we prefer to let the presentation of all compounds in this manuscript, as there are very few measurements in this high latitude zone.
There was no cross calibration between underway and Niskin samples. Therefore, following the reviewer's suggestion, we have added this note of caution in section 2.3:

"As no cross calibration was made between transect and Niskin measurements, possible differences between on-line and off-line measurements could not be evaluated. "

Specific comments

L18: please indicate months-year.
Done

L42: please support the "lifetime of 1 day" with a reference
The reference of Kloster et al. (2006) have been added

L70: this new sentence is a bit repetitive, as the previous already said oceans can be a sink for OVOCs.

We do not understand this comment, since this sentence shall highlight the fact that many OVOCs have a terrestrial source. We therefore like to keep this sentence.

L90: maybe Simó et al. (2022), which is cited in the Discussion, is an appropriate citation here
The citation has been added at the suggested place.

L111: please check the excitation emission wavelengths, they look very weird. Chlorophyll does not absorb much at 325 nm. Rather, it has an absorption peak in the blue, at around 430 nm, and fluoresces in the red, at around 680 nm.
We apologize for this. We now mention the correct values given by the company.
It reads now:

"wavelengths of 460 nm and 620-715 nm respectively"

L122: please specify material.
The material (glass) has been specified

L277, section 3.1.2: I suggest first mentioning the clear separation between bacterial communities from Atlantic and Polar water masses indicated by NMDS (Fig. 3).
We agree with the Reviewer's suggestion and have added the sentence
"Accordingly, communities markedly varied between Atlantic and polar waters >80°N (Fig. 3a)."

L474: the studies of Kettle and Lawso reported mean DMS/MeSH ratios of around 5.5. This corresponds to a MeSH / (DMS + MeSH) of 15%, higher than the <10% indicated by the authors.

≤15% is now given in the text

L486: cyanobacteria are well-known DMSP consumers, and are especially competitive under sunlight compared to heterotrophic bacteria. So It's not inconceivable that they produce MeSH as a DMSP assimilation by-product. Citations:
Malmstrom, R. R., et al. (2005). Dimethylsulfoniopropionate (DMSP) assimilation by Synechococcus in the Gulf of Mexico and northwest Atlantic Ocean. *Limnology and Oceanography*, *50*(6), 1924-1931.
Vila-Costa, et al. (2006). Dimethylsulfoniopropionate uptake by marine phytoplankton. *science*, *314*(5799), 652-654.
Ruiz-González, C., et al. (2012). Sunlight modulates the relative importance of heterotrophic bacteria and picophytoplankton in DMSP-sulphur uptake. *The ISME journal*, *6*(3), 650-659.

We agree that cyanobacteria can take up DMSP, and have modified the text as follows including the citation to Vila-Costa et al.:
" The link between cyanobacteria and MeSH potentially relates to the known uptake of DMSP by *Synechococcus* and *Prochlorococcus* (Vila-Costa et al., 2006), although DMSP-utilizing genes are overall rare in cyanobacteria (Liu et al., 2018)."

Technical corrections and typos

L80: Please correct: "This impact of sea-ice has been shown to DMSP"

For clarification, it has been reworded to
"This influence of sea-ice has been shown for DMSP, DMS, isoprene, acetone and acetaldehyde in the Canadian Arctic (Galindo et al., 2014; Wohl et al., 2022; Galí et al., 2021)"

L196: please check writing

The repetition has been deleted, the sentence now reads:
Measurements were typically performed every 2.5 minutes, except between 61.1 °N to 65.3°N, where measurements were performed every 10 min for approximately 24 h, to scan a wider range of masses (m/z).

L201: allows
Done

L309: “for surface values” → “near the surface”
Done

L333-341: check writing

The paragraph now reads:

“DMS and Chl a were strongly correlated (R^2 Pearson’s correlation coefficient = 0.93; Fig. S6). Isoprene also correlated with Chl a (R^2 = 0.6, Fig. S6) but only when excluding station 19. This correlation supports a biological source of isoprene, in agreement with previous demonstrated links between isoprene and Chl a maxima (Tran et al., 2013). Station 19 was the only station where diatoms almost exclusively dominated the phytoplankton biomass. The little emission of isoprene by cold-water diatoms (Bonsang et al., 2010) could explain this pattern.

In contrast to the latitudinal transect, MeSH showed low concentrations at most ice stations, with the exception of station 19. Station 19 was special due to its location above the shelf, and the phytoplankton community dominated by diatoms.

CO concentrations overall decreased with depth (Tran et al., 2013), except for station 31 with a CO peak at 30 m depth. “

Reviewer 2

Follow-up on Specific Comments

Line 201: How the MeSH sensitivity was determined presents uncertainty to the MeSH measurements, as the authors mention. Calculating the sensitivity as the average of sensitivities of compounds with similar m/z assumes that the sensitivity is primarily dependent on mass transmission. The authors state that the value used (13.4 ncps/ppb) is at the high range of measured calibration coefficients. It would be good to state what this range in calibration coefficients is. Other work that also has not had an experimentally measured calibration factor for MeSH has used the sensitivity to DMS given their similar collision rate constants and transmission efficiencies (Lawson et al. ACP (2020)). Another paper that measured calibration factors for MeSH and for DMS on a PTR-MS found that they were 3x more sensitive to DMS than MeSH (Novak et al. ACP (2022)). Since the MeSH findings in this paper are so important and have implications for the significance of MeSH in the sulfur budget, it would be good to have a more nuanced discussion of the uncertainty in MeSH based on what the range in calibration factor could be. I suggest this section of the supplemental provides a range in [MeSH] based on applying the average sensitivity of acetone and acetaldehyde, the DMS sensitivity, and the range in sensitivity coefficients in this study.

To follow the reviewer’s suggestion, we have now included the following paragraph in the supplement.

“As MeSH quantification has been performed by using a sensitivity coefficient based on the average of the sensitivity of acetaldehyde and acetone, the uncertainty associated to this choice has been estimated. For this, the estimated coefficient has been compared on one hand to an averaged sensitivity coefficient and on the other hand to an estimated DMS sensitivity coefficient. For the first comparison, the average sensitivity coefficient of 9.4 ncps/ppb represents the mean of 10 sensitivity coefficient (determined for m/z 33, 42, 45, 59, 69, 71, 73, 79, 93 and 107). If we would apply such a sensitivity coefficient to MeSH, it would increase all concentration by a factor of 1,43. So for example, the mean value of MeSH for polar water would be 4,24 nM instead of 2, 96 nM. As mentioned, DMS was not in included in the standard that we had at that time. Recently, we have purchased a NPL (National Physics Laboratory, Teddington, UK) standard containing a series a compounds, including acetaldehyde, acetone and DMS. We have performed 3 calibrations (on different days) at the laboratory with the same PTRMS used during the TRANSSIZ campaign. By taking into account the ratio of the DMS sensitivity (14, 4 ncps/ppb) compared to an average of acetaldehyde-acetone sensitivity (21,2 ncps/ppb), we obtained a value of 1, 47, so very close from the first evaluation. Finally, we conclude that due to the absence of a calibrated standard for MeSH, concentrations reported in this paper could be underestimated by a factor of 1.5.”

Line 202: Please provide information on how exactly this uncertainty was estimated. For example, what is the uncertainty in your calibrations, etc. and how does that lead to an overall value of +/- 20 or 30%. The provided reference Baudic et al. (2016) also does not provide this detail on how the uncertainty was calculated. This uncertainty estimation seems low given the lack of calibration to MeSH.

Information is now provided in Supplement S4. It reads as follows

"A complete estimation on the gas-phase measurement of this PTRMS has been performed in Baudic et al. (2016). This estimation, based on the ACTRIS measurement guidelines VOC 2014 (see Debevec et al., 2017), calculates the expanded uncertainty of U (X) as

$U(X) = k \times u(X) + DL_x/3$

With k being the coverage factor (here 2), u (X) the combined uncertainty in X, and DL_x *the detection limit of the species X. The combined uncertainty includes errors on standard gas, calibrations, blanks, reproducibility/repeatability, linearity, and relative humidity parameters. This expanded uncertainty has a maximum of 21% (21%, 18%, 9% and 10% for m/z 42, 45, 59 and 69 respectively). We do not give here the detailed contribution of each factor, as those calculations were not done specifically for this campaign. Nevertheless, we note that the two main sources contributing to the overall uncertainty were due to linearity error and to the uncertainty of VOC concentrations in the calibration standard gas. An additional uncertainty is the conversion of gaseous ppb into nM (based on the error of the calibration linearity, see calibration example for acetone in S4a). The overall uncertainty was then estimated at 21%, 32%, 11% and 11% for m/z 42, 45, 59 and 69 respectively. Therefore, the uncertainty for the calibrated compounds (including DMS, which was not present in the gas-phase standard but which has been calibrated with a liquid standard) has been estimated at about 30%.*

As MeSH has been quantified using a sensitivity coefficient based on the average of the sensitivity of acetaldehyde and acetone, we assessed the uncertainty by comparing (i) to an averaged sensitivity coefficient, and (ii) to an estimated DMS sensitivity coefficient. For (i), the average sensitivity coefficient of 9.4 ncps/ppb represents the mean of 10 sensitivity coefficients (determined for m/z 33, 42, 45, 59, 69, 71, 73, 79, 93 and 107). If applying such a sensitivity coefficient to MeSH, it would increase concentrations by a factor of 1.43. As mentioned, DMS was not present in the standard that we had at that time. Recently, we have purchased a NPL (National Physics Laboratory, Teddington, UK) standard containing a series of compounds, including acetaldehyde, acetone and DMS. We have performed three calibrations (on different days) in the laboratory with the same PTRMS used during TRANSSIZ. Taking into account the ratio of the DMS sensitivity (14.4 ncps/ppb) compared to an average of acetaldehyde-acetone sensitivity (21.2 ncps/ppb) we obtained a value of 1.47, hence almost identical to the first evaluation. Overall, we conclude that due to the absence of a calibrated standard for MeSH, concentrations reported in this paper could be underestimated by a factor of ~1.5."

We add here an information for the reviewer. New uncertainties estimations have been made for this PTRMS, in the framework of its long-term use at the ACTRIS-SIRTA station. These estimations, following updated guidelines from ACTRIS (which will be soon available), are described in detail in Simon et al. (2022), https://doi.org/10.5194/essd-2022-406. The overall uncertainty (taken into account precision and accuracy) has been estimated at 16%, 18%, 14% and 21% for m/z 42, 45, 59 and 69 respectively, consistent with the previous estimation.

The reviewer is right to point out a different uncertainty for MeSH. This has now been specified in the supplement (see above) and in the main text:

"The overall uncertainty for dissolved VOC measurements was estimated at ± 30%, except for MeSH. Due to the missing direct calibration of MeSH, its concentration could be underestimated by up to 1.5 times (see S4). Therefore, reported concentrations presented here for MeSH have to be considered as lower limit."

Line 363: I suggest moving the MeSH/(MeSH+DMS) values in Table 1 to a separate column. Also please clarify the unit for MeSH/(MeSH+DMS). This looks like percentage, if so, please add that to the column title.

A new column has been added to present MeSH/(MeSH+DMS) and the unit (%) has been added