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

Reviewer 3

The manuscript by Gros et al. reports on a large-scale survey of DMS, methanethiol and several other gases along the gradient between the eastern subpolar North Atlantic and the Arctic. Their main goal was to assess the concentrations and spatial distribution of these compounds and relate them to the environmental (physical, microbiological) conditions via statistical correlations. Based on their results, the authors aim to improve our current understanding on trace gas cycling in the context of a changing Arctic.

General assessment

The topic of this manuscript is certainly relevant for a wide biogeochemistry community and contributes to amend the large gaps of data coverage for marine trace gases; in particular for compounds which are understudied in comparison with other climate-relevant gases such as CO2 or CH4. The paper is very well written, its structure is clear and the methodological approaches are both sound and explained with enough detail. Although the manuscript is quite descriptive, the authors state clearly that they aim to report on the results of their survey. Hence, from that perspective, they were successful in achieving that. In my opinion a significant drawback of the study is the absence of air-sea fluxes of the different compounds measured in surface waters. Based on the content of the methods presented by the authors I cannot judge whether they have information at hand to do so, but it would be worth making an effort to provide estimates of air-sea fluxes for at least some of the compounds (I do think this can be done for DMS and CO).

We thank the reviewer for the overall positive evaluation of our manuscript and for the useful comments and suggestions. We agree that it would be very interesting to assess air-sea fluxes for the measured compounds and indeed our initial aims was also to perform measurements in the air, but it turned out that the parallel installed sampling line for atmospheric measurements aboard the vessel, dedicated for CO_2 measurements, had shown large contaminations. Thus, we decided to focus on ocean measurements only and in addition, we like to point out that we are here mainly focusing on the link between measured VOCs and biology (phytoplankton and bacteria). We think that adding some “theoretical” estimates of air-sea fluxes would go beyond the scope of this manuscript.

Other than that, most of my comments to the paper are minor (see below).

Specific comments

Title: The current title is rather long and can be misleading. The word “Variability” is should not be used generally here since the authors are addressing the spatial variability of several trace gasses, not their temporal variability.

The title is indeed quite long, but we do think it is important to mention the main elements. Following the suggestion of reviewer 2, we have added “dissolved” to the title, to avoid any confusion with atmospheric measurements. To address the suggestion of reviewer 3, we have now replaced “Variability” by “Concentrations”:

“Concentrations of dissolved dimethyl sulphide (DMS), methanethiol and other trace gases in context of microbial communities from the temperate Atlantic to the Arctic Ocean”.

29-30: Revise syntax, in particular after “understanding of”. Also, I recommend stating how specifically the paper contributes to that understanding, since right now this could mean anything.

We changed the sentence as follows:

Overall, the demonstrated latitudinal and vertical patterns contribute to understanding how concentrations of central marine trace gases are linked with chemical, and biological parameters across oceanic waters.

80: Please replace the word “levels” for “concentrations” here and it all instances where you refer to that quantity.

Done

81: “numbers” is too unspecific. Please refer precisely to which measurable quantity you are referring to here.

We agree and have changed to *“concentrations”*

82: Bacterial diversity and water masses were addressed. No sea ice data cover was presented and therefore it should not be presented as a factor in your experimental design.

Sea ice cover was removed

88: The citation to Peeken (2016) is unnecessary. I know such reports have a doi number, but they do not constitute a source of peer-reviewed information and should only used when absolutely needed (see journal's regulations)

The reviewer is correct that this reference is a report, but since it is the cruise report with valuable information about the expedition, we would prefer to keep it in the manuscript.

92: Stating that "usually" there is not sensor drift is not enough, even if the sentence is supported by a publication. The authors need to show that this was indeed the case during their survey in order to keep the credibility of their observations.

We do understand that the wording was a little misleading but please note, these tests are routinely carried out by the staff of Polarstern and we are only informed if the sensors are not behaving in the designated ranges. We thus wrote now:

"The instrument performs a self-cleaning routine every day with acid washing and freshwater rinsing. In addition, sensor behavior is controlled by staff members of Polarstern (for details see Petersen (2014))".

106-107: Here the citation is also unnecessary and should be removed. If the authors want to refer to the data used, there are better ways such a data set doi from Pangaea.

We disagree with the reviewer to delete this citation, since it credits the hard work people invest to create data products, which can be used by everybody if they need it for their studies. To credit the other colleagues involved in the CTD work we would like to keep this citation.

117: Delete point

Done

123: delete “l” after Chl a

Done

123: The R2 value in S1 is different than the one shown here. Revise.

The value given in S1 is R (0.91), the value given in the text is R2 (which have been changed from 0.84 to 0.83).

153-154: Revise wording. I would suggest “The measurement principle of PTRMS is (...)” or similar.

Done

214-217: The details on how this statistical analysis was setup should be explained in the “Material and Methods” section (i.e. independent and dependent variables, etc.). Otherwise the statement seems arbitrary (i.e. coming from nowhere).

We have added more information at the end of the bacterial method part:

“Nonmetric multidimensional scaling was performed to determine bacterial community variability along the transect. Associations between the abundance of bacterial ASVs and environmental parameters were determined via Holm-corrected Spearman’s correlations. Only correlations >|0.4| were considered, and only if higher than with latitude to omit indirect signals due to geographical variability”

234-235: Explain the details on how the system was adjusted. This reads as if the authors used the continuous system for profiling. Was that the case? If so, a detailed description is needed.

During the transect, continuous measurements were performed; for the vertical profiling, measurements were made on samples collected using the CTD rosette. This has been described in the section 2.1 (see below)

“Along the ship track between May 19th and 27th, trace gases were continuously measured in the surface water layer. (…).After May 27th, eight ice stations (number 19, 27, 31, 32, 39, 43, 46, and 47, Table S1) were carried out (...) During the ice stations, discrete seawater samples for trace gas and phytoplankton composition analysis were collected at six different depths of the water column using the CTD (conductivity, temperature, depth) water-sampling carousel. These samples were collected in 1 L light-proof flasks for direct analysis on board.”

As the reviewer refers to a sentence (line 234-235) which corresponds to the “results section”, we do not think this experimental information should be given here. Nevertheless, for clarity we now refer to section 2.1.

259-266: I am not convinced of the approach here. Why was station 19 removed from the analysis? It appears that although stations 19 and 32 have high productivity, both isoprene and CO behave completely different. Also, if one compares CO concentration at stations 19 and 39 (having contrasting chl a concentrations), it becomes evident that CO is not affected by the same processes as other gases. The reasons for this are unfortunately not discussed at all. In order to explain the variability of some of the CO concentrations at depth (e.g. at stations 32 and 43), the authors claim that differences in the profiles are due to “decreased photochemical production following lower light penetration”. However, this is the case for all stations and therefore it is not a compelling reason to explain the decrease with depth. Perhaps the authors rather refer to the effect of different sea ice coverage percentages in light penetration (?). If so, they can easily explore this possibility by using such data which is widely available.

We are very sorry, but we made a mistake when transferring the data from Excel to Sigmaplot for the final Fig 4 of the CO profiles, which was not measured at every station. Thus our results and discussion did not match completely the figure. We hope that our discussion about CO is now easier to follow. In addition we now clearly separate isoprene and CO in the discussion.

We also had the rationale of excluding station 19 for the correlation between isoprene and Chl a, since this is the only station where diatoms exclusively dominated the biomass. From our previous laboratory studies we did find that cold water diatoms emit less isoprene compared to temperate species (Bonsang et al. 2010) and thus we decided it was reasonable to exclude this station. This is now discussed as follows:

“Isoprene also markedly correlated with Chl a (R^2 = 0.6, Fig. S8), but only when excluding station 19. This correlation supports a biological source of isoprene, in line with the shown linkage of isoprene and Chl a maxima (Tran et al., 2013). Station 19 was the only station where diatoms dominated almost exclusively the phytoplankton biomass. As shown in laboratory

experiments, cold-water diatoms only emit little isoprene (Bonsang et al. 2010), which could explain the observed behavior.

In contrast to the latitudinal transect, MeSH showed low concentrations at most ice stations, except for station 19 (with higher concentrations and a clear decrease with depth). Station 19 was special since being located above the shelf and harbouring a diatom-dominated phytoplankton community it might be speculated that the diatom community also produces MeSH, but overall we have currently no real explanation as to why it is associated with a higher MeSH than the other stations.

The vertical profile of CO shows a decrease with depth as shown in Tran et al. (2013). This supports the notion that CO photoproduction (the main source of CO in the ocean) decreases up to threefold from the surface to 20 m depth (Fichot and Miller, 2010). An exception is station 31 where CO peaked at 30 m depth. This could indicate the presence of a large CO emitter, as the emission of CO can vary by more than an order of magnitude between phytoplankton species (Gros et al., 2009).

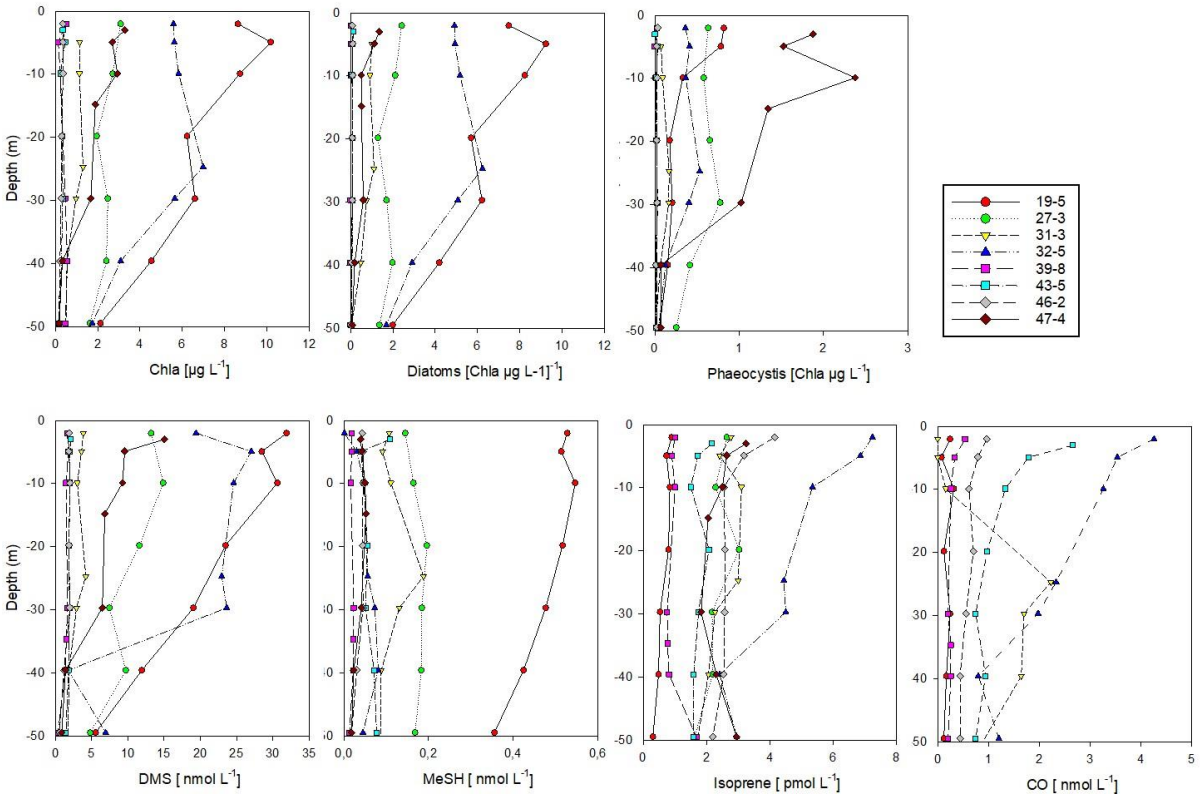

Fig. 4. *Biological parameters and trace gas vertical distribution (0-50 m depth) at sea-ice covered stations north of 80°. According to Dybwad et al., (2021) stations 39, 43 and 46 (Yermak Plateau) were in a pre-bloom phase, while all other stations were in a bloom phase. Stations 19 and 32 were shelf stations. The contribution of each phytoplankton group is expressed as Chl a concentrations.*

303: Is the mean value for surface waters or does it include the water column measurements? Please clarify.

It has been clarified as follows:

"For polar waters, the mean value of 5.9 ± 2.9 nM *in surface measurements during the transect* (...)"

308: Personal communications are not appropriate. Even less in this case since there are already two citations supporting the statement.

The personal communication has been removed

353: Same comment to personal communications. The authors already used Dybwad et al. (2021) as a defining criterion for the bloom stages in the study area at the time of sampling.

The personal communication has been removed. Instead we cite as suggested:

"Dybwad et al. (2021)"

385-387: This statement is contradictory with the results presented by the authors for CO. Based on the data presented it is only clear that CO production is not necessarily tied to a biological component and that photochemistry might have had a more significant role at the time of sampling. The authors argue (L.313-315) that low CO production by diatoms might be the explanation for the low concentrations at e.g. 19. However. this is speculative and cannot be substantiated with their observations. I recommend revising this aspect of the discussion.

We agree with the reviewer that the sentence "these probably have phytoplankton driven origins" does not apply to CO. Therefore, we have completed the sentence as :

Whereas isoprene, acetone, acetaldehyde and acetonitrile concentrations decreased northwards, CO, DMS and MeSH were uncorrelated with latitude and retained considerable concentrations in polar waters. Hence, these probably have phytoplankton-driven origins with regional variability, e.g. through localized blooms *and/or the presence of sea-ice.*

Supplementary information: there are inconsistencies in the naming of Figs. S6-S8. For instance, in L.235 S7 is mentioned although it does not match what is actually shown.

We are sorry about the confusion with numbering of figures S6 to S8. This has now been corrected.

In Fig. S8 no CO is shown (although announced in the main text) and the caption does not match the figure.

Indeed, this is a mistake in the main text, which should not mention CO when mentioning S8 (now S7). This has now been corrected. The figure caption of S8 (now S7) does correspond to the graphics (DMS and isoprene correlations with Chl a). As the figures S6 to S8 have now been correctly numbered, there should be no more confusion.