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

Rewiever 1

The paper by Gros et al. makes a substantial contribution to our understanding of the spatial distribution of climatically important trace gases and their potential underlying drivers. I would highlight the finding of very high DMS and significant MeSH concentrations under ice, the uncoupled DMS and MeSH distributions, and the distinct correlations between each VOC and bacterial ASVs at quite fine taxonomic resolution. Strong relationships between MeSH and bacterial ASVs in comparison to other VOCs is indicative of widespread bacterial DMSP metabolism. The paper is clearly written and well structured, and its messages are well supported by the observations. In the specific comments below I make some small criticisms that should be addressed. I suggest several additional citations, which I find important to both support the authors' findings and give fair credit to previous studies. I was also a bit disappointed by the little use authors make of HPLC data. My feeling is that they are missing an opportunity to assess the relative importance (even if based only on correlation patterns) of phytoplankton vs. heterotrophic bacterial diversity in controlling VOCs distribution in the area north of 80N that they sampled from Niskin bottles. My main criticism is with regards to the figures: they should be improved to make them more self-descriptive.

We thank the reviewer for the overall positive evaluation of our manuscript and for the useful comments and suggestions. We agree that it would have been great to link phytoplankton and bacterial diversity. However, please note that these data are unfortunately not fully comparable - HPLC & phytoplankton data are available for the depth profiles, while bacteria were only assessed in surface seawater. This prevents statistical comparisons. Please note, we have modified the figures according to your suggestions.

- Specific comments

- L39: Please add more up-to-date references, given that major advances in understanding of DMSP catabolism pathways have been made since 2007.
- L41: suggested citation: Kiene, 1996. Production of methanethiol from dimethylsulfoniopropionate in marine surface waters
- These articles may be of interest to provide a more complete view:
- L45-48: Rodríguez-Ros et al. 2020. Distribution and drivers of marine isoprene

- concentration across the Southern Ocean
- L49-50: Fichot and Miller, 2010. An approach to quantify depth-resolved marine photochemical fluxes using remote sensing: Application to carbon monoxide (CO) photoproduction
- L56: Acetaldehyde is also photoproduced: Zhu and Kieber, 2020. Global Model for Depth-Dependent Carbonyl Photochemical Production Rates in Seawater.
- L65: Lewis and Arrigo, 2020. Changes in phytoplankton concentration, not sea ice, now drive increased Arctic Ocean primary production
- L65: Galindo et al. 2014. Biological and physical processes influencing sea ice, under-ice algae, and dimethylsulfoniopropionate during spring in the Canadian archipelago
- L65: Wohl et al. 2022. Sea ice concentration impacts dissolved organic gases in the Canadian Arctic
- L66: Galí et al. 2019. Decadal increase in Arctic dimethylsulfide emission
- L68: Two good examples of changing phytoplankton species distribution
- Oziel et al. 2020. Faster Atlantic currents drive poleward expansion of temperate phytoplankton in the Arctic Ocean
- Orkney et al. 2020. Bio-optical evidence for increasing Phaeocystis dominance in the Barents Sea

We thank the reviewer for these suggestions of references, which have all been included in the introduction. In addition, we now also mention the new review on DMS including polar data https://doi.org/10.3390/microorganisms10081581 as well as the review of the microbiology of isoprene in aquatic system (https://doi.org/10.3354/ame01972)

- L259-260: I don't see how the correlation between DMS and Chl is connected to diatoms being the main photosynthetic group. Please rephrase.

- We apologize for this phrase, which indeed is misleading. The new text reads: "*We observed a strong correlation between DMS and Chl a (R-squared Pearson's correlation coefficient = 0.93; Fig. S8). Since diatoms were the most prominent photosynthetic group at ice-covered stations (Fig. 4) we consider them important for DMS fluxes in the Polar Ocean.*"

- L261: I cannot see the cyan squares of station 43 in the CO panel.

Thank you for spotting this, we had mixed up the columns for CO when transferring it from Excel to Sigmaplot for the final version. Please find a new Fig. 4 with the correct CO profiles, which were not carried out at every station.

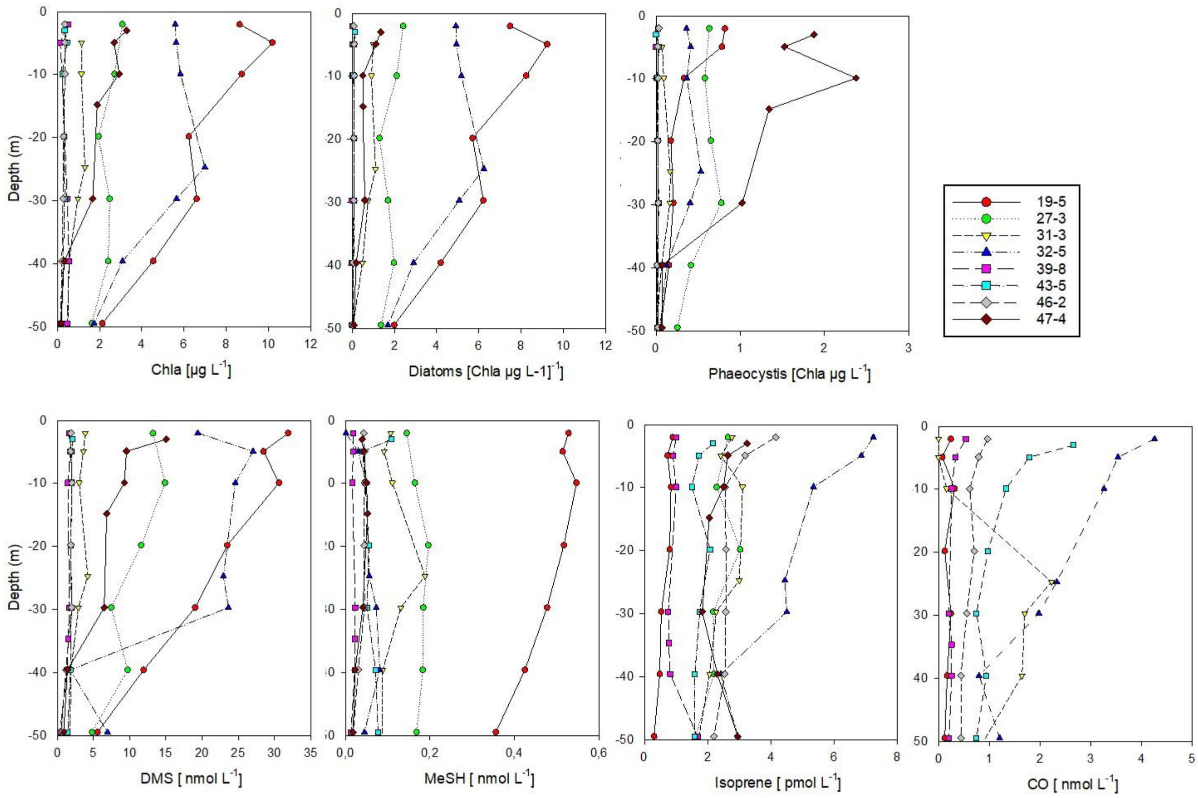


Fig. 4. *Biological parameters and trace gas vertical distribution (0-50 m depth) at sea-ice covered stations north of 80°. According to Dybwad et al., (2021) stations 39, 43 and 46 (Yermak Plateau) were in a pre-bloom phase, while all other stations were in a bloom phase. Stations 19 and 32 were shelf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

- L305: Perhaps mention that Arctic waters feature much higher CDOM content than typical oceanic waters. The approach used by Conte et al. (2019) estimated CDOM using a biooptical relationship between CDOM and Chl developed for typical (case I) oceanic waters (Morel et al., 2009). Arctic waters do not conform to this bio-optical type and are typically seen as optically complex waters with compound influence of oceanic, riverine and icemelt waters with distinct signatures in terms of CDOM and particle loads. Failing to account for the high CDOM content is likely to result in underestimation of CO photoproduction.

  We thank the reviewer for this clarification and changed the text accordingly:

  "*Elevated values in the Arctic are not reproduced by the NEMO-PISCES model (Conte et al., 2019), which might be caused by the bio-optical relationship between CDOM and Chl-a. This model was originally developed for typical oceanic waters (Morel & Gentilli., 2009). However, Arctic waters do not conform to this bio-optical type and are typically considered optically complex waters, with distinct signatures of CDOM and particle loads through the influence of oceanic, riverine and ice-melt waters (Goncalves-Araujo et al. 2018). Conte et* *al., 2019 attribute the release of CO and/or CDOM to sea-ice melt or to lower bacterial consumption in cold waters. The first hypothesis is supported by up to 100 nM CO measured in sea ice (Xie and Gosselin, 2005; Song et al., 2011)."*

- L332: A note of caution: the values reported by Davie-Martin et al. (2020) for the NAAMES expedition are not credible in the case of DMS production rates. The highest DMS production they found in May, around 43 nM h-1 (1000 nM d-1), is about 15 times higher than any previous measurement (Galí and Simó, 2015, their figure 3a). This might have been caused by the bubbling in their experimental setup, which is known to induce DMS production in stressed cells (eg Wolfe et al., 2002. Dimethylsulfoniopropionate cleavage by marine phytoplankton in response to mechanical, chemical, or dark stress). Similar artifacts may have affected measurements of the production rate of other VOCs in that study.

  We thank the reviewer for this note of caution. Concerning the statement about acetonitrile for which oceans can be a small source or sink, we have replaced the citation of Davie-Martin et al. (2020) by Singh et al. (2003) and Williams et al. (2004). We have nevertheless left this citation to underline the potential microbial utilization of acetone and acetonitrile.

- L349: Suggested citation: Hayashida et al. 2020. Spatiotemporal Variability in Modeled Bottom Ice and Sea Surface Dimethylsulfide Concentrations and Fluxes in the Arctic During 1979 – 2015. Quoting from their abstract: "...model results indicate that the bottom ice DMS and its precursor dimethylsulfoniopropionate production can be the only local source of oceanic DMS emissions into the atmosphere during May prior to pelagic blooms".

  The citation of Hayashida has been added, in addition to Levasseur, 2013.

- L350-351: this view can be nuanced. At high latitudes, the seasonal correlation between DMS and Chl is typically positive and quite high. See e.g. Lana et al. (2012) Reexamination of global emerging patterns of ocean DMS concentration, their fig. 4. Also:

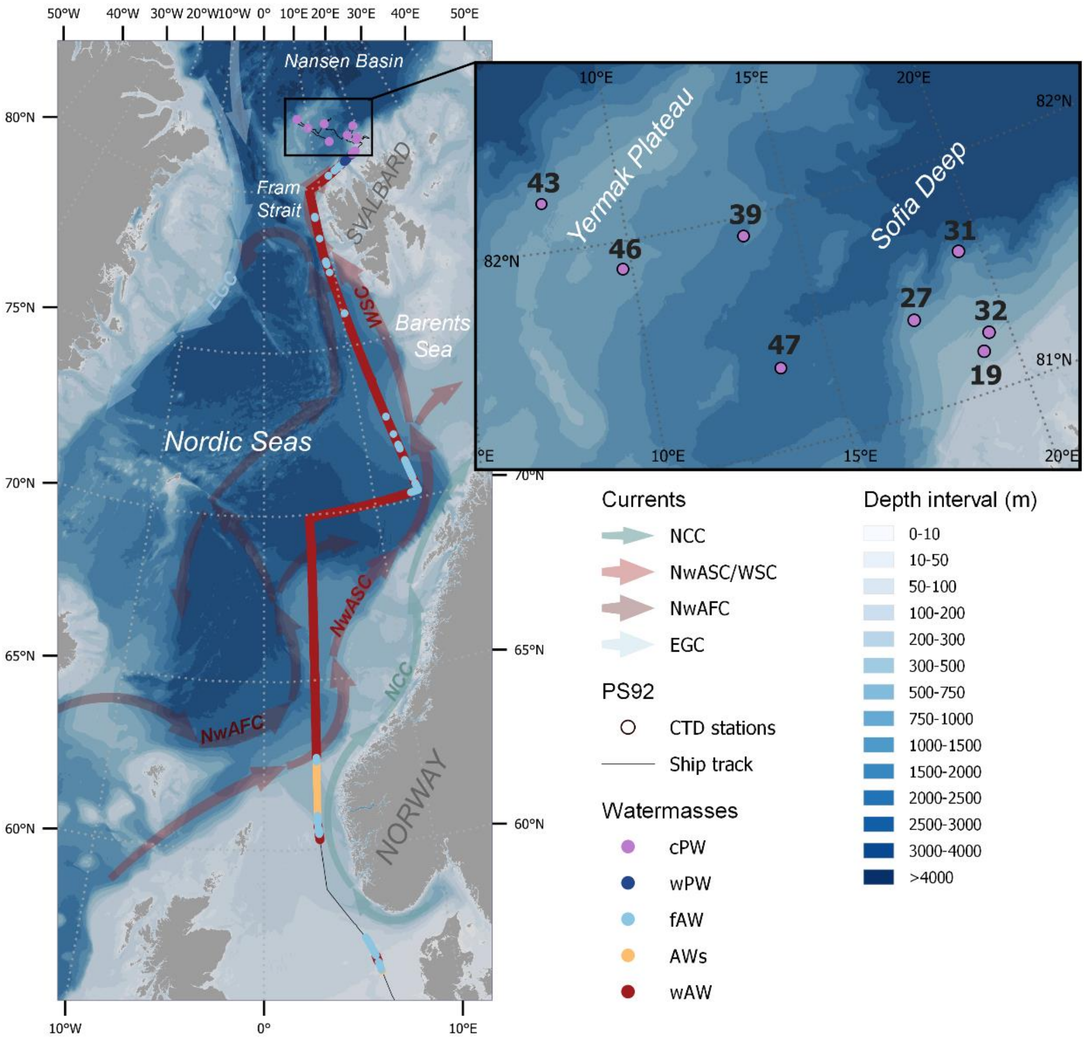
- Galí et al., 2018. Sea-surface dimethylsulfide (DMS) concentration from satellite data at global and regional scales. Table 1 and Fig. 7.
- Wang et al., 2020. Global ocean dimethyl sulfide climatology estimated from observations and an artificial neural network. Table 1, section 3.

We thank the reviewer for this comment. We have nuanced this view in the revised version, now reading as follow :

"Stefels et al. (2007) have suggested no direct relationship between DMS and Chl a on global scale, since the precursor of DMS (DMSP) is produced by diverse phytoplankton at different rates, connected to their physiological state. However, different approaches employed by Gali et al. (2018) and Wang et al. (2020) have shown that Chl a can be a strong predictor of DMS concentrations. In addition, Lana et al. (2012) reported that the DMS-Chl a correlation strongly varies with latitude, with a positive correlation at high latitudes (north of 40°N and south of 40°S). Nevertheless, we note that the figure presented by Lana et al. (2012) shows a lower correlation on the region covered by our transect. The poor correlation found along our transect (R^2 =0.1) probably reflects different phytoplankton types and bloom stages.

- It would be very helpful to see the main currents and the different water masses on this map. For example, this would support the description given in L180-186, several parts of the Results and Discussion, the summary given in Table 1, etc.

We thank the reviewer for this suggestion, which was also suggested by reviewer 2. We have modified Figure1 (and caption) to now include the main surface currents (arrows) as well as the water masses (colored dots) along the ship track (the latter was earlier shown in Fig S6). It also includes a legend for the bathymetry (background blue colors). The new figure is perhaps a little busy, but has the benefit of including all relevant information in one figure, better supporting several parts of the manuscript. Fig S6 has thus been omitted.



NEW Fig. 1: The ship track with the sampled range colored by water mass: 'regular' warm Atlantic Water (wAW), coastal influenced Atlantic water with low salinity (AWs), freshened and cooled Atlantic Water (fAW), warm Polar Water (wPW) and cold Polar Water (wPW), according to the temperature and salinity criteria in Table 1. Surface measurements were sampled by the FerryBox system between 57°N and 81°N, while vertical profiles were sampled by a CTD rosette at eight sea ice stations (black insert and Table S1). The background map shows the GEBCO_2022 bathymetry (GEBCO Compilation Group (2022); doi:10.5285/e0f0bb80-ab44-2739-e053-6c86abc0289c) and a schematic overview of the major currents influencing the surface waters in the study area, as adopted from Skagseth et al. (2022): the northward flowing Norwegian Atlantic Slope Current (NwASC), West Spitsbergen Current (WSC), Norwegian Atlantic Front Current (NwAFC), and Norwegian Coastal Current (NCC) and the southward East Greenland Current (EGC).

The current-related text in the manuscript (section3.1) was also slightly modified with the aim to better describe the sampled water masses along the ship track:

"Along the latitudinal transect, we performed online surface measurements of Chl a and hydrographic parameters, covering five different water masses: warm Atlantic Water with low salinity (AWs), 'regular' warm Atlantic Water (wAW), freshened and cooled Atlantic Water (fAW), cold Polar Water (cPW) and warm Polar Water (wPW) as defined in Table 1 following Tran et al. (2013). The major part of the

transect, from 63 to 80°N, occurred in wAW (see Fig. 1). Fresher (low-saline) Atlantic Water (AWs and fAW) was encountered in the vicinity of the Norwegian Coastal Current (NCC) which carries water masses influenced by river run-off: AWs at 60.6-62.3 °N (with fAW in the mixing zones), and fAW around 70-72°N. Fresher mixed products (fAW) were also intermittently encountered west of Svalbard (where AW meets fjord/coastal water masses), as well as in the marginal ice zone where AW mixes with and gradually subducts under fPW. Polar Water (PW) only occurred north of 80°N in the Nansen Basin."

- Figure 2: I strongly recommend to depict somehow the water masses along the transect, for example with colored horizontal bars on top of the plot.

We thank the reviewer for this idea, which was also suggested by reviewer 2. The revised figure includes now as a first panel a horizontal bar representing the different water masses.

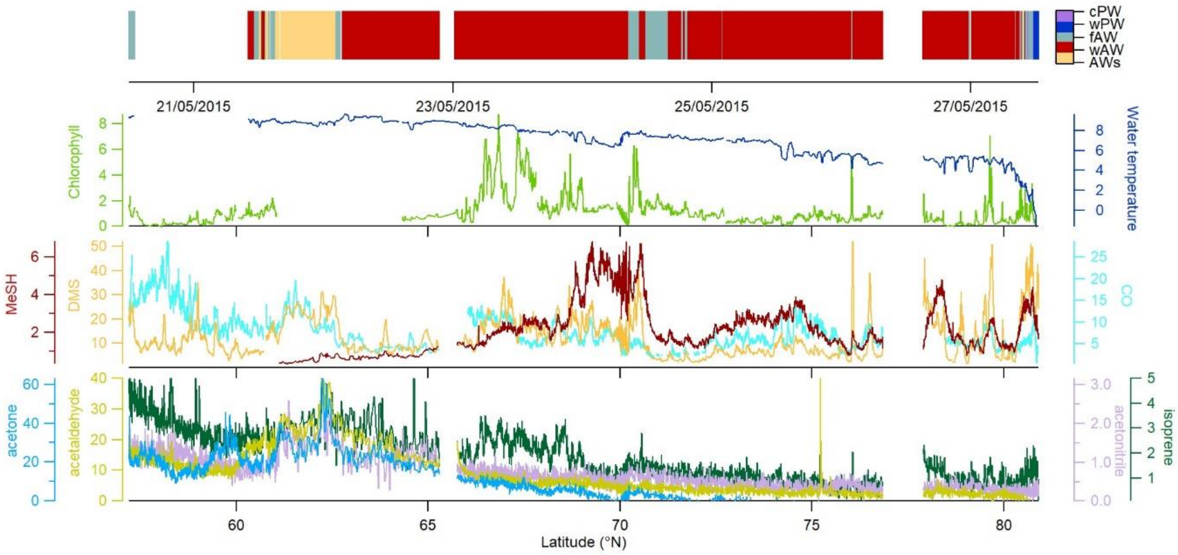


Figure 2: Latitudinal variability of acetone (nM), acetaldehyde (nM), acetonitrile (nM), isoprene (pM), DMS (nM), MeSH (nM), and CO (nM) between 57.2°N to 80.9°N in relation to Chl a (µg L^{-1}) and water temperature (°C). Due to sensor failure temperature values are missing until ~61°N. The top X-scale gives the corresponding date. On the top panel, the colored horizontal bar represents the different water masses as in Figure1 and Table 1

- Figure 3: It would be useful to provide more commonly used taxonomic classifications/levels for some bacterial genera. For example, Yoonia-Loktanella and Ascidiaceihabitans tell nothing to me, but I immediately associate Rhodobacteraceae with certain types of reduced sulfur metabolism.

In the discussion, Yoonia-Loktanella and Ascidiaceihabitans are specified as Rhodobacteraceae and how this family commonly performs sulfur cycling. Rhodobacteraceae are also mentioned in the abstract. We think that this figure would become difficult to read if more taxonomic info were added to each ASV, and would therefore prefer to leave it as is.

- Figure 4: Please indicate (for example in the legend) whether stations are in pre-bloom or bloom stage, perhaps distinguishing the shelf stations as well.

Thank you for this suggestion, the new figure captions reads:

*"**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."*

- Figure 5: same as Fig. 2.

As for Fig. 2, we have tested plotting the horizontal bar with the water masses classification, but found that it does not add meaningful information. Hence, we have decided to leave the figure as is.

- Technical corrections and typos
- L156: "at their" repeated
- L254: "but the here found concentrations" sounds a bit awkward, please reword
- L292: "as already been found", please remove "been"
- L324: nM, not nm

All technical corrections have been done