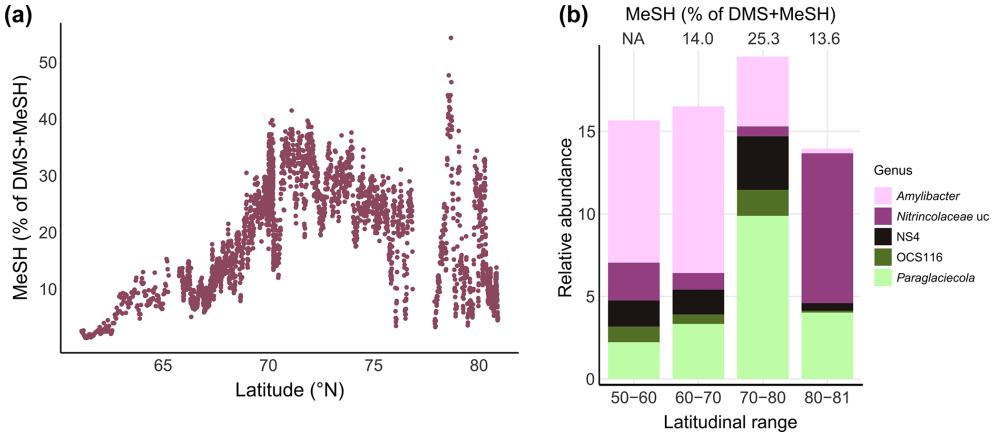

Figure 5. MeSH contribution to the sulfur budget and associated bacterial patterns. **(a)** Latitudinal variation of the MeSH fraction in relation to the total concentration of DMS + MeSH. **(b)** Relative abundance of selected bacterial genera by latitudinal range, , with the corresponding MeSH / DMS ratio displayed on top CE9. NA: not available; uc: unclassified. TS3

2022). In pelagic waters, DMS generally dominates gaseous sulfur, with MeSH being the second most abundant compound contributing on average $\leq 15\,\%$ to the total sulfur species in the North and Baltic seas (Leck and Rodhe, 1991), in the Atlantic Ocean (Kettle et al., 2001) and in the southwest Pacific Ocean (Lawson et al., 2020).

Comparable to some North Sea locations (Leck and Rodhe, 1991), MeSH contributed up to 40 % between 70–75° N in our study, with a maximum of 50 % at 78.6° N (Fig. 5a). This latitudinal variability was underlined by shifts in major bacterial genera. For instance, *Paraglaciecola* (Gammaproteobacteria) and NS4 (Bacteroidetes) peaked together with the highest MeSH fraction between 70–80° N. Abundances of *Amylibacter* decreased towards the north, whereas unclassified *Nitrincolaceae* prevailed north of 80° N together with an again smaller MeSH / DMS ratio (Fig. 5b). The overall MeSH contribution of 20 % suggests that MeSH represents a considerable fraction of sulfur, with linkages to microbial dynamics. Accordingly, we found several correlations with the abundance of specific ASVs. Correlations between *Yoonia-Loktanella* and *Ascidiaceihabitans* ASVs with MeSH reflect the prominent role of *Rhodobacteraceae* in DMSP demethylation (Curson et al., 2011; Moran et al., 2012). The positive relation of SAR11 and SUP05 ASVs corresponds to the prevalence of DMSP-metabolizing genes in these taxa (Nowinski et al., 2019; Landa et al., 2019; Sun et al., 2016). 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). Overall, these observations indicate yet undescribed chemical linkages among primary producers.

5 Conclusion

We present the first measurements of DMS, MeSH and other trace gases along a transect from the North Atlantic to the ice-covered Arctic Ocean. High-resolution latitudinal data between 57 and 80° N were complemented with vertical profiles at sea-ice stations north of 80° N. 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 likely have phytoplankton-driven origins with regional variability, e.g. through localized blooms and/or the presence of sea ice. The DMS peak in polar waters pointed to sea ice as reservoir of DMS (Levasseur, 2013) and the prevalence of DMS-emitting phytoplankton. The marked correlation between DMS and Chl *a* in the diatom-dominated region north of 80° N represented a typical marginal sea-ice zone effect. The missing correlation between DMS and MeSH suggested different processes of production and degradation, although both compounds originate from DMSP. Although DMS was overall more abundant, MeSH contributed on average 20 % (and up to 50 %) to the total DMS + MeSH budget, suggesting consideration of MeSH as a secondary aerosol producer in some regions. The potential importance of MeSH was underlined by more and stronger bacterial correlations than with DMS, indicating that bacterial DMSP demethylation is important across extensive latitudinal gradients. Notably, higher acetaldehyde concentrations north of 80° N suggest that ice-covered regions could be a reservoir of acetaldehyde. While artefacts from off-line measurements (sampling through Niskin bottles) cannot be completely excluded, this result indicates a potential role of this reactive compound in regional atmospheric chemistry. However, a comprehensive understanding of marine trace gas dynamics, including the rapidly changing Arctic, requires further measurements in seawater, sea ice

Proof only