



Nitrous oxide and methane in a changing Arctic Ocean

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Abstract Human activities are changing the Arctic environment at an unprecedented rate resulting in rapid warming, freshening, sea ice retreat and ocean acidification of the Arctic Ocean. Trace gases such as nitrous oxide (N_2O) and methane (CH_4) play important roles in both the atmospheric reactivity and radiative budget of the Arctic and thus have a high potential to influence the region's climate. However, little is known about how these rapid physical and chemical changes will impact the emissions of major climate-relevant trace gases from the Arctic Ocean. The combined consequences of these stressors present a complex combination of environmental changes which might impact on trace gas production and their subsequent release to the Arctic atmosphere. Here we present our current understanding of nitrous oxide and methane cycling in the Arctic Ocean and its relevance for regional and global atmosphere and climate and offer our thoughts on how this might change over coming decades.

Keywords Arctic Ocean · Environmental change · Ice retreat · Methane · Nitrous oxide · Ocean acidification · Warming

BACKGROUND

The Earth's polar regions are rapidly changing as a direct result of our altered climate. The recent IPCC special report on the oceans and the cryosphere (IPCC 2019) found that “The polar regions are losing ice, and their oceans are changing rapidly. The consequences of this polar transition

extend to the whole planet, and are affecting people in multiple ways”. The changes that are being experienced are having and will continue to have an increasing effect on the biogeochemical processes which are fundamental to the functioning of marine ecosystems (Rees 2012). The Arctic Ocean (AO) is experiencing this climate driven modification of its environment faster than anywhere else on the globe and there is high confidence that this region will likely become practically sea ice-free during the seasonal sea ice minimum for the first time before 2050, the practically ice-free state is projected to occur more often and with higher greenhouse gas concentrations (Fox-Kemper et al. 2021). To add to the complexity of this change, the loss of ice will be accompanied by the combined effects of increasing temperatures (Huang et al. 2017) and ocean acidification (AMAP 2018). Each of these artefacts of change have the potential to disrupt the biological processes which control the production and consumption of the atmospherically important trace gases nitrous oxide (N_2O) and methane (CH_4), whilst some of these effects may also modify the physical exchange between ocean layers or between the ocean and atmosphere.

As both of these gases are strongly radiatively active and their release from the Arctic Ocean will be impacted by ongoing and projected environmental changes, there is a need to further our understanding of how this might alter regional and global climate and ultimately impact society. The project “Pathways and emissions of climate-relevant trace gases in a changing Arctic Ocean (PETRA)” was designed to address these issues. In this paper we set observational and experimental evidence from a PETRA research cruise to the Fram Strait in July 2018 on board RV Polarstern (PS114) within the context of previous measurements of N_2O and CH_4 made by ourselves and others over the pan-Arctic region to assess our current

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understanding and to consider likely scenarios of change of N_2O and CH_4 in a future Arctic Ocean.

NITROUS OXIDE (N_2O)

N_2O is a long-lived atmospheric trace gas whose atmospheric mixing ratio is increasing at a mean rate of 0.85 ± 0.03 ppb yr^{-1} (Canadell et al. 2021). It is rated as the third most important greenhouse gas (GHG) in the troposphere (Butler and Montzka 2018; Canadell et al. 2021) with a global warming potential on a 100-year timescale of approximately 300 times that of CO_2 (Ramaswamy et al. 2001; Etmann et al. 2016) and is involved in ozone (O_3) depletion in the stratosphere (Ravishankara et al. 2009). Though the N_2O concentration in most of the surface of the ocean is in close equilibrium with the atmosphere (Nevison et al. 1995), global emissions from the open ocean and coastal waters contribute 35–39% of the total natural sources of N_2O (Tian et al. 2020) and there is a fine balance between the ocean acting as net producer or consumer of N_2O . Environmental effects associated with a changing climate, which include rising temperatures, oxygen depletion and ocean acidification are quite likely to impact the level of this equilibrium (Bange et al. 2019).

Nitrous oxide is biologically produced through three processes: Denitrification is the anaerobic reduction of NO_3^- to N_2 which has N_2O as an intermediate; nitrification involves the two stage aerobic oxidation of NH_4^+ through NO_2^- to NO_3^- , where the release of N_2O as a by-product is dependent on the ambient O_2 concentration (Goreau et al. 1980; Löscher et al. 2012). In the third route, nitrifier-denitrification, N_2O can be formed during the reduction of NO_2^- via nitric oxide to N_2O by ammonia oxidizing bacteria. The pathway by which N_2O is produced by ammonia oxidizing archaea is not yet fully understood (Wu et al. 2020).

METHANE (CH_4)

CH_4 is the most abundant organic trace gas in the environment, it plays an important role in the Earth's climate and as a result of anthropogenic activities its atmospheric mixing ratio has more than doubled since the preindustrial era (Etmann et al. 2016). CH_4 acts to limit the tropospheric oxidative capacity and is the second most important greenhouse gas, with a global warming potential that exceeds CO_2 by up to 32 times over a 100-year timescale (Etmann et al. 2016; Canadell et al. 2021), contributing approximately 20% of the radiative climate forcing for all GHGs. The world's oceans are a natural source of CH_4 but play only a minor role in its global atmospheric budget, the

open ocean and coastal waters account for 7 to 12% of the total natural sources and approximately 4% of global emissions (Saunois et al. 2020). It is thought that the open ocean contributes only a minor proportion of atmospheric CH_4 with coastal environments including estuaries thought to account for approximately 75% of the total marine source (Weber et al. 2019).

The origin of CH_4 in marine waters is from a diverse range of sources which may be geological, including from hydrothermal vents, cold seeps, and CH_4 clathrates, or microbial in origin. The source of CH_4 in ocean waters continues to prove enigmatic. Traditional understanding suggests that microbial methanogenesis is an anaerobic process, which in oceanic waters is thought to occur either in oxygen depleted waters or in anoxic micro-environments that are associated with zooplankton guts and particulate material (Brooks et al. 1981; Bianchi et al. 1992; Marty et al. 1997). However, the enhanced CH_4 surface saturations found far away from shelf areas are difficult to explain because conventional CH_4 production via archaeal methanogenesis should not occur in the well-oxygenated surface waters. This “ocean methane paradox” has been explained in tropical and sub-tropical oligotrophic conditions by the decomposition of methylphosphonate in phosphorus starved conditions (Karl et al. 2008) whereas in Arctic waters an alternative in situ CH_4 production has been proposed following the microbial cleavage of dimethylsulphoniopropionate (DMSP) (Damm et al. 2008, 2010, 2015). Indeed DMSP is often found in very high concentrations in the AO during sea ice algal blooms and sea ice brines, and is projected to increase following ongoing changes to the Arctic environment (Campen et al. 2021).

CURRENT UNDERSTANDING OF N_2O AND CH_4 IN ARCTIC WATERS

Concentrations of dissolved N_2O in the surface waters of Arctic shelf areas and central deep basins which exchange with the atmosphere are remarkably variable and can range from pronounced undersaturation to high supersaturation (Table 1). In general, ice-free surface waters appear to be undersaturated with N_2O whereas ice-covered surface waters are supersaturated with N_2O (Kitidis et al. 2010; Randall et al. 2012; Fenwick et al. 2017). For the North American AO it was suggested that high levels of N_2O were largely associated with production in the shelf sediments of Chukchi and Bering Sea's with subsequent advection eastwards (Fenwick et al. 2017). Supersaturations of N_2O were also associated with the continental shelf for this region during the 7th Chinese National Arctic Research Expedition (Zhan et al. 2017, 2021) who also

Table 1 Exemplar N₂O and CH₄ publications from Arctic waters to indicate variability in regional source-sink characteristics for the two gases in the upper water column likely to exchange with the overlying atmosphere

Region	Saturation Range (%) (Concentration Range (nM))	Potential for Source - Sink	Reference
N₂O			
Labrador Sea – South Baffin Bay	109 ± 9 (15.4 ± 1.6)	Equilibrium-Source	(Kitidis et al. 2010) <100% attributed to melt-water. >100% found under multi-year sea-ice
Baffin Bay – Marginal ice	117 ± 24 (19.8 ± 3.1)	Source	
Baffin Bay North & Lancaster Sound	116 ± 12 (17.6 ± 1.6)	Source	
Canadian Archipelago	117 ± 15 (21.0 ± 2.8)	Source	
Amundsen Gulf – Beaufort Sea	115 ± 7 (17.0 ± 1.0)	Source	
Amundsen Gulf: Sea Ice	~15 – 39 (<3 – 7.9)	Sink	(Randall et al. 2012)
Water under ice	60 – 111 (11 – 18.8)	Sink	
Chukchi Sea: Inner shelf	106 – 157 (12.1 – 25.1)	Source	(Hirota et al. 2009, Zhan et al. 2017, Zhan et al. 2021) (Zhang et al. 2015) (Zhang et al. 2015, Zhan et al. 2021)
Outer shelf	95 – 100 (12.7 – 16.4)	Equilibrium-Sink	
Abyssal plain	92 – 94 (16.3 – 16.9)	Sink	
Bering Sea	102 – 137 (9.6 – 21.8)	Source	(Hirota et al. 2009, Zhan et al. 2017, Zhan et al. 2021)
North American Arctic:	77 – 145 (10.9 – 24.6)		(Fenwick et al. 2017)
Surface waters – ice free	<100%	Sink	
Surface waters – ice covered	<110% (<17.8)	Source	
Alaskan Coastal Water	~100%	Equilibrium	
Pacific Summer Water	<120% (mean 17.0)	Source	
Pacific Winter Water	<145% (mean 24.6)	Source	
Eurasian basin: Nansen Basin	42 – <100	Sink	(Verdugo et al. 2016)
Amundsen Basin	<100 – 111	Source	
Nordic Seas	82 – 100 (9.7 – 15.1)	Sink	(Zhan et al. 2016, Rees et al. 2021) This study, Fig. 1a
	95 – 107 (11.6 – 18.2)	Sink to Source	
CH₄			
Labrador Sea – South Baffin Bay	139 ± 45 (4.2 ± 1.3)	Equilibrium-Source	(Kitidis et al. 2010)
Baffin Bay – Marginal ice	170 ± 51 (5.9 ± 1.6)	Source	
Baffin Bay North & Lancaster Sound	164 ± 65 (5.3 ± 2.1)	Equilibrium-Source	
Canadian Archipelago	226 ± 94 (8.2 ± 3.5)	Source	
Amundsen Gulf – Beaufort Sea	151 ± 76 (4.9 ± 2.8)	Equilibrium-Source	
North American Arctic:	95 – 220 (3.0 – 7.3)	Equilibrium-Source	(Fenwick et al. 2017) (Li et al. 2017)
Surface Bering, Chukchi, Canada Basin.	97 – 517 (3.5 – 20.9)		
Surface Canadian Arctic Archipelago	<420% (mean 15.0)	Source	(Fenwick et al. 2017)
Central Arctic:			(Damm et al. 2010)
Atlantic Water	(1.5 – 3.5)	~Equilibrium	
Pacific Water	(~4 – ~6)	Source	
Fram Strait:			(Damm et al. 2015)
Atlantic Water	(3 – 3.5)	~Equilibrium	
Pacific Water	(4 – 9)	Source	
Nordic Seas	93 – 132	Sink to Source	This study, Fig. 1b

noted that offshore waters tended to be in equilibrium with the atmosphere. The Nordic Seas associated with the Greenland Basin and Fram Strait have been reported to be undersaturated in N₂O (Rees et al. 2021) and act as a permanent sink to atmospheric N₂O (Zhan et al. 2016).

During research cruise PS114 onboard the German vessel RV *Polarstern* to the Fram Strait in July 2018 we measured concentrations and saturations of dissolved N₂O and CH₄ (Fig. 1) using an autonomous equilibrator headspace setup coupled to a trace gas cavity ringdown spectroscopy

analyzer (Picarro Inc., USA; see Supplementary Material for methodology). The saturation of a dissolved gas indicates the relative amount of gas held relative to the value expected (100%) when the water body is in equilibrium with the overlying atmosphere. Supersaturated waters (> 100%) indicate a local source of the particular gas whereas a value of undersaturation (< 100%) indicates the potential for a sink to the atmosphere. In Fig. 1a, saturations of N₂O can be seen to vary from a minimum of 95% in the southeast of the region close to Svalbard associated with the poleward West Spitsbergen Current and Atlantic influenced open-water. In the west of the region, higher levels of N₂O, up to 107%, were found in shallower, ice-covered water close to Greenland and associated with the southerly flowing East Greenland Current. The heterogeneous distribution of N₂O does not relate directly to surface ice-cover and maybe associated with the origin of source waters, though the higher concentrations to the west are likely the product of shelf sediment production and limited exchange with the atmosphere.

At present, N₂O emission estimates indicate that the overall net release of N₂O from the AO to the atmosphere is comparably low (Fenwick et al. 2017). Microbial nitrification and/or denitrification in shelf sediments as well as water column nitrification have been proposed as the main N₂O production pathways in the AO shelf areas and central deep basins (Kitidis et al. 2010; Verdugo et al. 2016; Fenwick et al. 2017), whereas loss processes include biological consumption (Verdugo et al. 2016; Rees et al. 2021) and physical advection (Zhan et al. 2016). Indications for the future release of N₂O are conflicting. It was suggested that the ongoing decline of the Arctic sea ice cover may enhance future N₂O emissions to the atmosphere (Kitidis et al. 2010) whilst in contrast, ongoing ocean acidification (AMAP 2018) has been shown to decrease N₂O production in AO waters (Rees et al. 2016). The dominating N₂O pathways and their dependence from changing environmental parameters/stressors (temperature, ocean acidification) and modifications of exchange across boundary layers need to be verified. Improved emission estimates of N₂O to the atmosphere, as well as mechanistic understanding on how they might be affected by the above-mentioned perturbations is crucial since currently the uncertainties on the marine source of this gas to the atmosphere in the region are extremely high at approximately 100% (Yang et al. 2020).

Very high CH₄ surface saturations have been observed in the shelf areas and the central deep basins of the AO (Table 1). Vast areas of the AO seafloor, particularly those associated with Siberian, Chukchi, and Beaufort Seas, is rich in permafrost (a potential substrate for methanogenesis) and CH₄ hydrates (Chen et al. 2021). At present, the AO is a potentially important source of atmospheric CH₄

(Kort et al. 2012) with sedimentary production via methanogenesis, dissociating gas hydrates and diffusion from geological dissolution providing the dominant source in shelf sea areas. Release of bubbles from sedimentary origin during the melting season represents a considerable, yet spatially constrained, source to the atmosphere (Thornton et al. 2020), though there is evidence to indicate that CH₄ released at the sea bed may not reach the atmosphere, e.g. (Myhre et al. 2016). In Fig. 1b, CH₄ saturations in the Fram Strait region during July 2018 can be seen to be highly variable. As with the distribution of N₂O (Fig. 1a) and patterns of heterogeneity previously reported, minimum saturations (~ 93%) of CH₄ were associated with Atlantic influenced ice-free waters and the highest observed (132%) were found in ice-covered areas close to the Greenland coast suggesting a strong source of CH₄ to the air for this region.

Future CH₄ emissions from open ocean regions of the AO will largely be determined by aerobic CH₄ oxidation in the water column and 'non-conventional' microbial CH₄ production via DMSP (Damm et al. 2010, 2015). The increased supply of organic matter from rivers and permafrost thaw may further enhance microbial methanogenesis. The effect of future environmental stressors such as warming and pH on aerobic CH₄ oxidation is largely unknown (James et al. 2016).

EVIDENCE FOR POTENTIAL CHANGE

Impact of warming

Warming conditions are likely to impart direct and indirect effects on the processes controlling production and consumption of both gases. Whereas the solubility of gases is governed by well constrained laws of physics under the control of temperature and salinity, so that warmer waters hold less gas than cold ones, the composition of microbial communities and their ecological function of changing systems is much less predictable. Warming would further enhance microbial methanogenesis with a 2° increase in temperature resulting in a 25–200% increase in methanogenesis (Bange et al. 1998). Finally, sea ice loss as a result of warming may result in shorter residence time for CH₄ and N₂O in the water and thereby evasion to the atmosphere rather than in-water microbial processing (sea ice is currently considered as a semi-permeable barrier to air-sea exchange) (Kitidis et al. 2010). Increasing seawater temperatures lower the solubility of CH₄ in seawater and allow a shallowing of the CH₄ hydrate stability zone. Therefore, a small increase in seawater temperature could potentially lead to hydrate dissolution and the subsequent release of CH₄ from the AO to the atmosphere (Kitidis 2009).

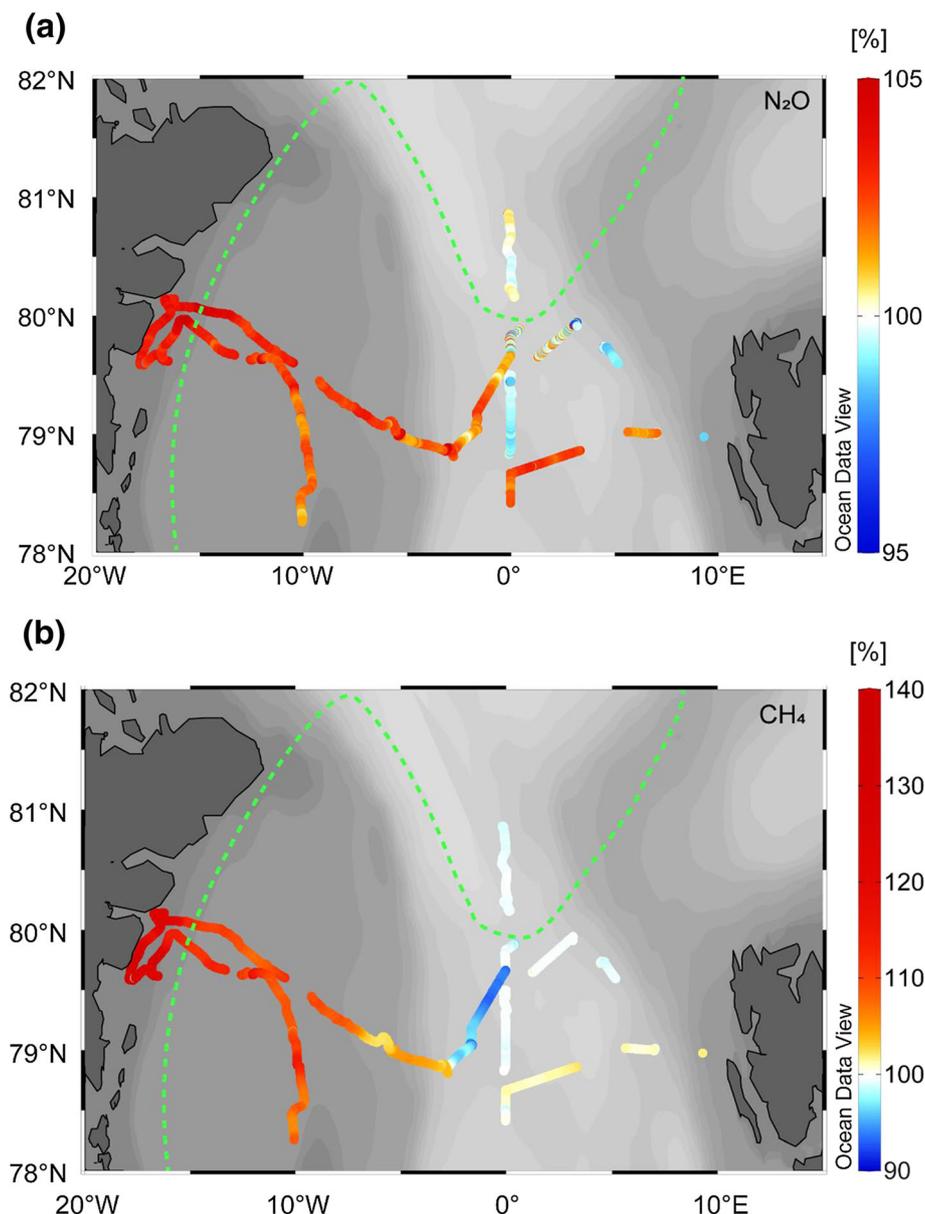


Fig. 1 Distribution of (a) N₂O and (b) CH₄ saturation during research cruise PS114 onboard the RV *Polarstern* during July 2018. The green dotted line in each image represents the southern extent of sea ice at 90% cover (see Supplementary Material for method description)

There is some confidence that phytoplankton primary productivity is likely to increase as ice-cover retreats (Lannuzel et al. 2020) which will increase the sediment load of organic material that is potentially used in remineralisation processes in the generation of both N₂O and CH₄. Increases in primary production can be attributed to the interplay between two factors: first, the increased spatial and temporal extent of open waters, and second, the enhanced nutrient input brought about by mixing, upwelling and lateral advection, all of which is fostered by increased inflow from subpolar seas (the so-called Atlantification of Arctic waters) and more frequent storm events (Polyakov et al. 2017).

As ocean temperatures rise, CH₄ hydrates may become unstable releasing vast quantities of CH₄ to the atmosphere which in turn may lead to further temperature increase and hydrate de-stabilisation according to the “clathrate gun” hypothesis (Kennett et al. 2003).

Although CH₄ solubility will decrease with increasing temperature, methanotrophy will also increase (Yvon-Durocher et al. 2014). Methanotrophy is the dominant oceanic CH₄ sink and is a first order process with respect to CH₄ concentration and inversely related to its turnover time. This suggests that an enhanced sedimentary release, potentially caused by warming triggered gas hydrate dissociation may result in shorter turnover times of dissolved

CH₄ in the water column (James et al. 2016). However, direct ebullition from dissociating gas hydrates or thawing permafrost may result in substantial emission of CH₄ to the atmosphere as bubbles rise faster than they are consumed by microbes (Shakhova et al. 2010). Decreased sea ice would also reduce the extent of areas where bubbles released from the sediments are trapped. The transfer of these bubbles across the sea surface during the melting season might represent a spatially variable, yet considerable source of CH₄ to the atmosphere (Zhou et al. 2014).

IMPACT OF OCEAN ACIDIFICATION

In the open oceans nitrification is the dominant mechanism for the production of N₂O. A number of studies have shown that nitrification rate is inhibited by decreasing pH (Beman et al. 2010; Kitidis et al. 2011) but the impact on N₂O appears equivocal. During experiments performed in Arctic and Antarctic waters (Rees et al. 2016) showed that whilst the microbial community of ammonia oxidising archaea (the dominant nitrifying organisms) seemed unaffected by changing pH, the production of N₂O decreased at all stations by between 2.4 and 44% when pH was reduced at between values of 0.06 and 0.4 pH units. The reduction in N₂O yield from nitrification was directly related to a decrease of between 28 and 67% in available NH₃ as a result of the pH driven shift in the NH₃:NH₄⁺ equilibrium. In the subarctic western North Pacific (Breider et al. 2019) found that decreasing the pH during experimental manipulations acted to significantly increase N₂O production, whilst rates of nitrification either remained stable or decreased, indicating a de-coupling of the two processes. The differences between the findings of Rees et al (2016) and Breider et al (2019) maybe attributable to regionally associated differences in N₂O production pathways, or to the relative sensitivities of microbial communities found in the two regions.

During the current project we performed four experiments during PS114 in the Fram Strait region similar to those described in Rees et al (2016). These were conducted to examine the impact of ocean acidification in isolation and in combination with warming of 2 °C (see Supplementary Material for methodology). Initial findings can be seen in Fig. 2. It would appear that there is a decrease in both N₂O concentration and in nitrification rate with changes of pH which were made to match future conditions indicated by representative concentration pathway (RCP) of 6.0 and 8.5. Whilst there is some inherent variability in the responses, it would appear that there is no obvious response of N₂O production or nitrification rate to warming of 2 °C.

To date we are unaware of experimental evidence to suggest that either methanogenesis or methanotrophy show

any sensitivity to changing conditions of ocean acidification. A limited number of experiments that we have performed have all indicated that both CH₄ production and consumption processes are likely to prove resilient to ocean acidification in the AO.

ECOSYSTEM MODELLING

The incorporation of up-to-date knowledge of N₂O and CH₄ dynamics in the marine environment into coupled physical-biogeochemical models is important to (a) expand our understanding and to test hypotheses related to the dynamics of these gases, (b) to explain spatial and temporal distribution patterns, and (c) to predict future change under the impact of multiple stressors. However there is currently insufficient understanding of the production and consumption pathways of these gases and their environmental controls which remains a limiting factor for their wider inclusion into process-based model studies, both in the AO and elsewhere.

A range of model formulations of N₂O dynamics based on observational data have been developed in recent years and applied at a global scale, e.g. (Martinez-Rey et al. 2015; Ji et al. 2018) thus covering, but not focusing on, the AO region. However, implementation of regional-scale models of N₂O is hindered by the scarcity of observational data and incompleteness in understanding of various pathways and their response to stressors, especially within high-gradient environments.

The issue is even more apparent for CH₄, as many unknowns related to its production and consumption pathways still limit the rare modelling efforts to sensitivity studies, e.g. (Wahlström and Meier 2014) who focused on the Laptev Sea region. Better understanding of the controls over the CH₄ pathways is critical to constrain process-based models: for instance, the reported range of oceanic aerobic CH₄ oxidation rates spans several orders of magnitude. In the AO the uncertainties in biological pathways are further augmented by a range of shelf sea processes related to permafrost thawing and CH₄ hydrate dissolution, that all require advancements in understanding and consideration in the models.

The implementation of sophisticated process-based models which are able to project future emissions of N₂O and CH₄ under the influence of multiple stressors is severely hampered by sparse sampling (Weber et al. 2019) and lack of experimental evidence to advance statistically sound mechanistic understanding of the controlling processes. There is real need for an increased capacity of measurements such as those reported in this study to further develop this on both regional and global scales.

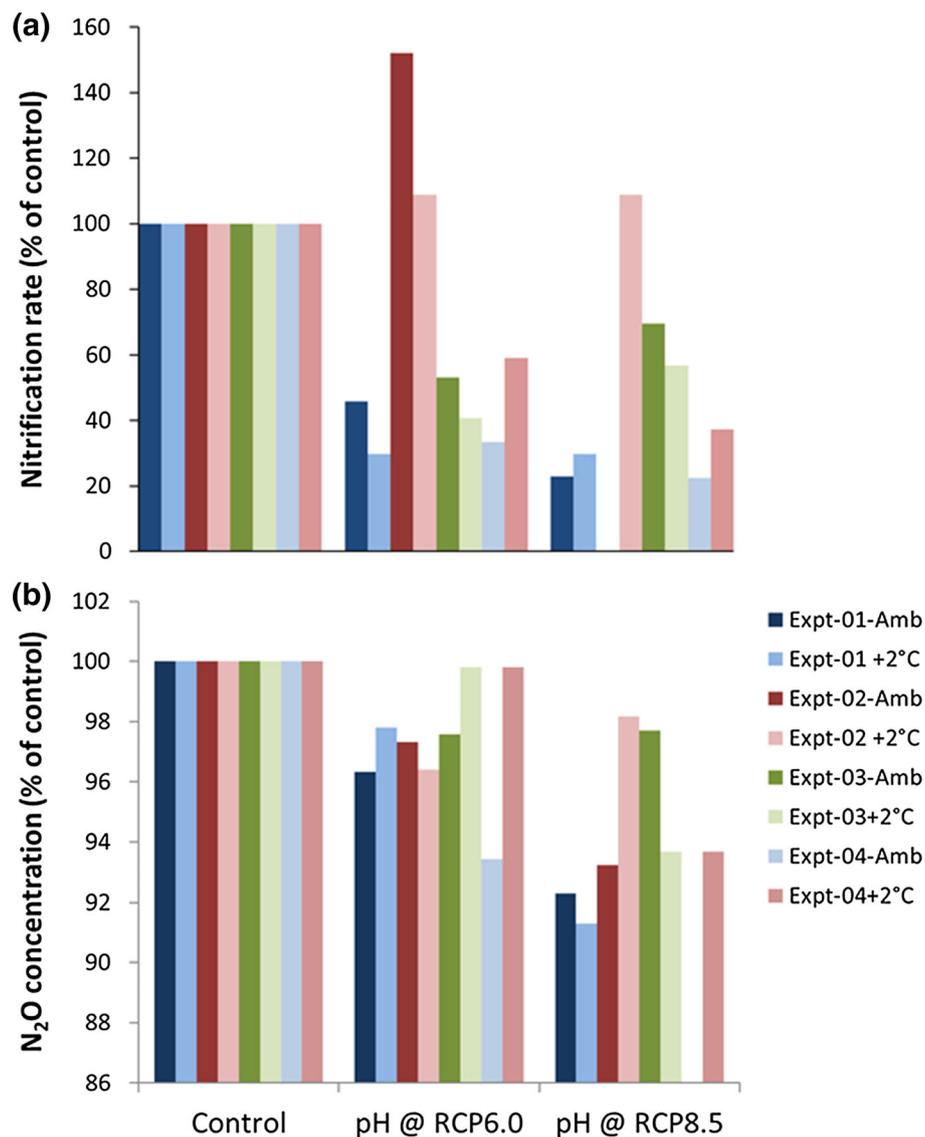


Fig. 2 The impact of decreasing ocean pH and increasing temperature by 2 °C on (a) the rate of nitrification and (b) N₂O concentration at four positions in the Fram Straits region of the AO during research cruise PS114 in July 2018 (see Supplementary Material for method description)

SOCIAL AND POLICY IMPLICATIONS

N₂O and CH₄ contribute significantly to climate change. They are relevant to the United Nations Framework Convention on Climate Change (UNFCCC), the primary international, intergovernmental forum for negotiating the global response to climate change. The ultimate objective of the UNFCCC is to stabilize GHG concentrations “at a level that would prevent dangerous anthropogenic (human induced) interference with the climate system” (UNFCCC 1992). The goal of the Paris Agreement (UN 2015), a legally binding international treaty on climate change, adopted by 196 Parties at UNFCCC COP 21 in Paris in 2015, is “to limit global warming to well below 2, preferably to 1.5 °C, compared to pre-industrial

levels”. To achieve this long-term temperature goal, countries aim to reach global peaking of GHG emissions as soon as possible to realize a climate neutral world by mid-century.

UN Member States adopted 17 Sustainable Development Goals (SDGs), as part of the 2030 Agenda for Sustainable Development (United Nations 2015), a global partnership for sustainable improvement of human lives whilst protecting the environment, including oceans, and tackling climate change. The oceanic production of N₂O and CH₄ addressed in this study is relevant to climate change goal (SDG13), to “Take urgent action to combat climate change and its impacts”.

The generation of impact is integral to PETRA (Fig. 3). Data generated will be managed and archived at our local

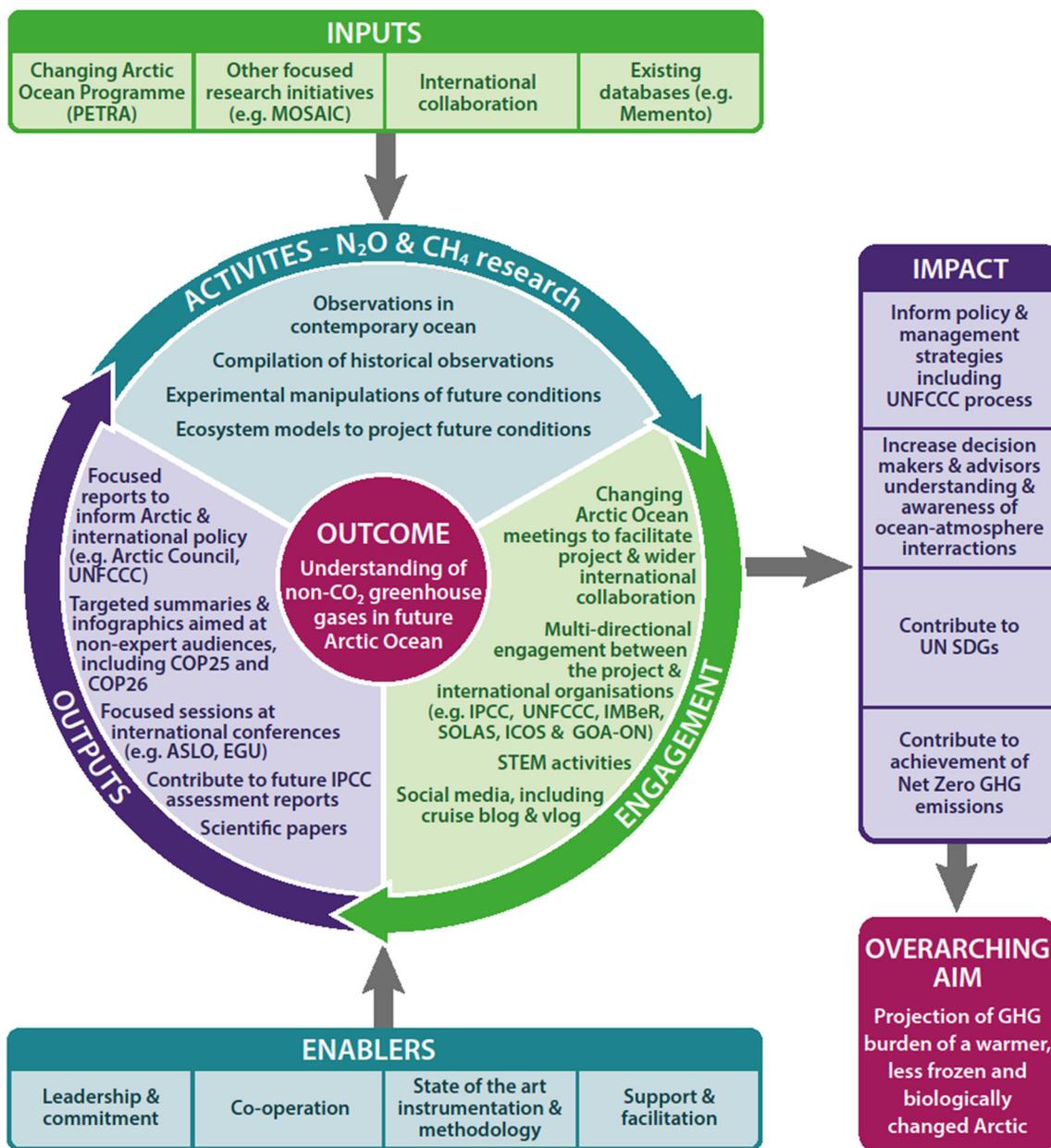


Fig. 3 Theory of Change visualisation of social and policy implications associated with the PETRA project investigations of N₂O and CH₄ in a changing AO

oceanographic data centres (British Oceanographic Data Centre for UK and PANGEA for Germany) according to our data management plan. To ensure greater visibility and access, CO₂ data collected during fieldwork expeditions in 2018 and 2019 have been submitted to the Surface Ocean CO₂ Atlas (SOCAT) and included in the annual Global Carbon Project budget (Friedlingstein et al. 2020) whilst N₂O and CH₄ data will be submitted to the marine methane and nitrous oxide database (MEMENTO) (<https://memento.geomar.de/>). Outputs will also be made available through engagement with the GOA-ON observational network.

“The polar regions are losing ice, and their oceans are changing rapidly. The consequences of this polar transition extend to the whole planet, and are affecting people in multiple ways” (IPCC 2019). Understanding the role of the ocean in the cycling and production of N₂O and CH₄ and how these may change in an ocean undergoing rapid and long-term change is therefore essential to the Convention and climate negotiations. The findings of our previous work (Rees et al. 2016) and the newer observations from PETRA indicate that ocean acidification has the potential to decrease N₂O emissions by up to 0.8 Tg N yr⁻¹ which is

Table 2 Sources and sinks of N₂O and CH₄ in the AO, the expected effect of ocean warming (and associated melting) and acidification, as well as the level of uncertainties in the current state of knowledge

Processes	Source/sink	Estimated overall effect ^a		Level of uncertainty
		Warming	Ocean acidification	
N₂O				
Nitrification	Source	↑	↓	Medium
Denitrification	Source/sink	↑	?	Medium
Air-sea exchange	Source/sink	↑	↓	High
Within-ice cycling & fluxes	Source/sink	?	?	Very high
CH₄				
Methanogenesis	Source	↑	?	Medium
Methanotrophy	Sink	↑	?	Medium
Aerobic oxidation	Sink	↑	?	High
Air-sea exchange	Source/sink	↑	?	High
Within-ice cycling & fluxes	Source/sink	?	?	Very high

^a↑ increase, ↓ decrease, ? unknown

comparable to all current N₂O production from fossil fuel combustion and industrial processes of 0.7 Tg N yr⁻¹. There is some potential therefore for decreases in N₂O release to the atmosphere to offer a negative feedback to global warming, though it is still too early to say as other contrasting (Breider et al. 2019) and compounding effects are still to be accounted for.

OUTLOOK

Whilst estimates of the global marine source of N₂O and CH₄ to the atmosphere have significantly improved (Wilson et al. 2020), a mechanistic understanding of the causes for the observed variability in sink-source dynamics and sea/ice-air gradients in the AO is missing. This degree of uncertainty is highlighted in Table 2, where current understanding of the processes controlling these gases in concert with ocean acidification and warming are presented. Reduced sea ice coverage in the AO and the adjacent subpolar regions with future warming will likely expose larger ocean surfaces to direct exchange with the atmosphere, increasing the overall source of N₂O and CH₄ (a positive feedback on GHG-driven warming). However, the sea air transfer depends strongly on the pre-existing gradients between both reservoirs. The balance is a complex product of several processes. During the freezing period, brine rejection leads to gas enrichment and density-driven fluxes towards the underlying water column, but also potentially supports temporary fluxes towards the atmosphere. In contrast, during melting, a dilution effect causes gas undersaturation with respect to atmospheric equilibrium together with a strong salinity driven stratification. This would result in a net ocean uptake unless a

strong source (e.g. sedimentary CH₄ release) or buoyancy-driven flow breaks the stratification causing deep mixing and upwelling. At present it is challenging to establish which process is dominant over the annual cycle since the cycling of N₂O and CH₄ within the sea ice is not well understood and local production can be masked by lateral advection. Moreover, it is not clear to what extent an ocean acidification driven change of N₂O production would contribute to offset the expected increase in the emissions to the atmosphere, and whether any synergistic effects may arise.

Resolving gradients across the sea–ice–air interfaces and their spatial and temporal variability requires a combination of dedicated, multidisciplinary surveys, time series observations and the use of novel methods and autonomous platforms suitable for different ice conditions, e.g. (Bange et al. 2019; Lee et al. 2019). Studies investigating dynamics of N₂O and CH₄ within sea ice are scarce and therefore should be addressed in future joint projects. Recent observations from the MOSAiC expedition (<https://mosaic-expedition.org/>) and the Synoptic Arctic Survey (<https://synopticarcticsurvey.w.uib.no/>) are expected to provide important contributions towards a better understanding of the cycling and emissions of N₂O and CH₄ in the AO. Overall, strengthening observational capabilities in the AO will reduce the current emission uncertainties and thereby improve our projections of future GHG emission trends within the context of global coupled models.

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