

***Interactive comment on “Ideas and perspectives:
A strategic assessment of methane and nitrous
oxide measurements in the marine environment”
by Samuel T. Wilson et al.***

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Reviewers comment»Line 42: reference should be 23, not 22

Authors response»Thank you for catching this typographic error – it has now been changed.

Reviewers comment»Line 56: It was good to see the “numerical modeling” portion in the abstract. And modeling came up throughout the review, but it might be more informative to have a section dedicated to what is needed for these models, in a comprehensive way. Specifically, what sort of temporal and spatial resolution is needed?

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What sort of precision on measurements is required?

Authors response»Reviewer #2 raises two important questions about the relationship between observations and models. Specifically Reviewer #2 queries (1) the level of temporal-spatial resolution and (2) the analytical uncertainty required to improve and constrain the models. Our response to these questions and instances where the text has been amended is provided below. A useful contextual analysis of analytical uncertainty associated with CH₄ and N₂O can be provided by consideration of discerning long term trends. The ocean's response to increasing atmospheric concentrations of CH₄ and N₂O can be discerned over a timescale of 10 and 5 years respectively, with an analytical uncertainty of 1% (assuming all other parameters remain equal). Stated another way, if we wish to determine whether the oceanic inventory of dissolved CH₄ and N₂O is increasing at the same rate as the atmosphere, we need to wait 10 years for CH₄ and 5 years for N₂O, with an analytical uncertainty of 1%. This topic was also discussed in Bange et al (2019) which stated 'Detecting inter-annual N₂O signals will require a precision of better than 0.02 nmol L⁻¹ (<0.2%)'. We agree with Reviewer #2 that it is important the manuscript reflects the need for high quality CH₄ and N₂O measurements and the text has been amended to specifically include this. Lines 526-531 now read 'Currently, there is no defined level of analytical uncertainty for CH₄ and N₂O analysis that would facilitate the establishment of 'high quality' measurements. However, attaining an analytical uncertainty of ≤1% is considered achievable and for context this would permit the ocean's response to the increasing tropospheric CH₄ and N₂O mole fractions to be resolved on timescales of 10 and 5 years, respectively, assuming all other parameters remain constant'. Achieving a <1% analytical uncertainty would facilitate more accurate inclusion of the mechanisms driving N₂O and CH₄ cycling in Earth System models, such as the relationship between N₂O yields and O₂ concentrations. However, care should be taken that the observations are providing the most useful data needed to improve the models. The manuscript already notes this by commenting that increased resolution of N₂O emissions in Earth System models would derive from greater constraint of the Michaelis-Menten kinetics associated with N₂O production as

a dependent of O₂ concentration. The manuscript text states in Section 7 that this could be achieved from laboratory based measurements where Lines 450-454 read 'Future model parameterizations for N₂O will require information on the variability of microbial process yields derived from culture studies with controlled varying conditions of O₂, pH, temperature, and nutrients'. The situation is different for the coastal environment which is one of the most uncertain and least predictable sources of methane and nitrous oxide. Using methane as an example, methane concentrations can vary by several orders of magnitude across spatial distances ranging from meters to kilometers. For example, Figure 5b shows methane concentrations increasing by at least 100-fold as depth decreases from 100 m to 5 m. In this setting, accumulating sufficient data points along coastal gradients to resolve the spatial distributions becomes a greater priority than achieving the highest possible analytical accuracy. We have amended the manuscript text to better reflect this and the legend for Figure 3 which illustrates the range of spatial-temporal phenomena that influences CH₄ and N₂O distributions now states on Lines 1226-1228 'The low resolution oceanographic surveys are more likely to achieve a high level of analytical accuracy compared to high resolution coastal measurements, however this is compensated for by high temporal resolution achieved by underway sampling'. Reviewer #2 also queries whether the manuscript should have a section for the modeling work. However, the preference of the authors is to discuss the insights from models and observations together in the context of the different science themes. One of the workshop objectives was to promote closer collaboration between modelers and observationalists in order to create more complementary tools to answer the most pressing scientific questions. Finally, we wish to point out that it is not just the analytical uncertainty in the CH₄ and N₂O measurements that requires improvement. As noted in the text on Lines 395-397 'a fivefold variation in CH₄ emissions from a single system occurred when applying different parameterizations to the measured gradients in CH₄ (Ferrón et al., 2007)'.

Reviewers comment»Line 141: check out: Gelesh, L., et al (2016). Methane concentrations increase in bottom waters during summertime anoxia in the highly eutrophic

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estuary, Chesapeake Bay, USA. *Limnology and Oceanography* 61, S253-S266.

Authors response»The manuscript already cites Gelesh et al (2016) in Section 5 ‘CH₄ and N₂O in shallow marine environments’. This is our preferred location for the reference rather than long-term time-series observations.

Reviewers comment»Line 172: can you be more specific on what predictor variables are for methane and what are for N₂O? Just separate the citations here for which gas they focus on and what they find are the predictors

Authors response»This has now been clarified and Lines 174-178 now read ‘Machine-learning mapping also recently identified the various contributions of physical and biogeochemical predictor variables for CH₄ (e.g. depth, primary production; Weber et al., 2019) and N₂O distributions (chlorophyll, sea surface temperature, apparent oxygen utilization, and mixed-layer depth; Yang et al., 2020).’

Reviewers comment»Line 190: “other processes”. Please elaborate on what processes you mean here.

Authors response»We apologize for the ambiguity associated with this sentence. The text has been revised and Lines 192-196 now state ‘In the surface waters of tropical and temperate oceans, a number of factors contribute to the low supersaturation of CH₄ including direct aerobic production arising from the degradation of methylated sulfur compounds by phytoplankton (Klitzsch et al., 2019) and methyl phosphonate in phosphorus-depleted waters (Karl et al. 2008, Sosa et al., 2020), indirect production via grazing (Schmale et al., 2018) and abiotic photoproduction (Li et al., 2020).

Reviewers comment»Line 197: check out: Lorenzen, T.D., Griener, J., and Coffin, R.B. (2016). Dissolved methane in the Beaufort Sea and the Arctic Ocean, 1992–2009; sources and atmospheric flux. *Limnology and Oceanography* 61, 300-323. And, Lapham, L., et al (2017). Dissolved methane concentrations in the water column and surface sediments of Hanna Shoal and Barrow Canyon, Northern Chukchi Sea. Deep-

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Authors response»The manuscript now includes the Lorenson et al. (2016) and the Lapham et al. (2017) references on Lines 204 and 205, respectively.

Reviewers comment»Line 220: check out: Lapham, L., et al (2013). Temporal variability of in situ methane concentrations in gas hydrate-bearing sediments near Bullseye Vent. *Geochemistry, Geophysics, Geosystems* 14, 2445-2459.

Authors response»Thank you for the suggestion, this reference has now been included.

Reviewers comment»Line 242: check out: Grant, N., J., and Whiticar, M.J. (2002). Stable carbon isotopic evidence for methane oxidation in plumes above Hydrate Ridge, Cascadia Oregon Margin. *Global biogeochemical cycles* 16 (4), 1-13.

Authors response»The Grant (2002) reference suggested by Reviewer #2 provides an in depth analysis of stable isotope methane values and concentrations to determine the quantitative fate of methane of entering water-column from the cold seeps of Hydrate Ridge. However, it doesn't include the broader analysis of higher order hydrocarbons which is the point of the text on Lines 246-249 'For example, combining these measurements with the ratio of CH₄ to higher order hydrocarbons (e.g. ethene (C₂H₄) and ethane (C₂H₆)) can be used to infer for example, whether the origin of the CH₄ is thermogenic, sub-seafloor, or biogenic within the water column'. The three references we have cited (Whiticar, 1999; Pohlman et al., 2009; Lan et al., 2019) all include the analysis of additional hydrocarbons in order to provide greater contextualization for the origin of methane.

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