Biogeosciences Discuss.,
https://doi.org/10.5194/bg-2020-270-AC1, 2020

Biogeosciences Discussions

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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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Received and published: 22 September 2020

Reviewer #1 The ocean is the net source of both CH4 and N2O, which are the second and third largest anthropogenic greenhouse gases. However, the air-sea flux of these gases remains uncertain, due mainly to the lack of sufficient reliable measurement of marine CH4 and N2O. It is thus of urgent need to further strengthen the observation of these greenhouse gases in the ocean. To this end, the authors proposed several perspectives of improving the current observation ability to better constrain and predict the marine CH4 and N2O flux. Overall, I feel these perspectives are essential and clearly stated. The manuscript has included the main findings of previous researches

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in this field.

Authors response »Thank you for your comments

Reviewer comment»My main concern of the manuscript is that the three initiatives been proposed here are quite similar with the main ideas of the recent study by the authors themselves (i.e., Wilson et al., An intercomparison of oceanic methane and nitrous oxide measurements, 2018; Bange et al., A harmonized nitrous oxide (N_2O) ocean observation network for the 21st Century, 2019).

Authors response »Reviewer #1 makes the comment that the three initiatives (development of SOPs, intercomparison of seawater samples, and improved usage and output for a centralized data repository) mentioned in Section 7 'Outlook and Priorities' have already been written about in two previous manuscripts. This is a valid comment with regards to the SOPs as their need was first articulated by Wilson et al (2018) and now two years later they are again being advocated for. In our defense, the SOP documents are being produced at this very moment. The work that is currently being undertaken is listed in response to Reviewer #1's next comment. We have now revised the third initiative that previously focused on improved use of a centralized data repository. The third initiative now highlights the need for a Global Data Product for CH_4 and N_2O. At the moment, the MEMENTO data repository collects CH_4 and N_2O concentrations which are then used by the modeling community. This activity has been very successful, but it occurs without the publication of any Data Product which would represent a quality controlled synthesis of all the concentration data that have been collected to that point. The absence of a Global Data Product impedes the progress of community-driven CH_4 and N_2O research on several levels as scientists measuring CH_4 and N_2O do not receive the appropriate acknowledgement for use of their datasets in Earth system models and there is no common Data Product for the modeling community to use. This situation for CH_4 and N_2O contrasts sharply with that of pCO_2 which releases Global Data Products on an annual basis via Surface Ocean CO_2 Atlas initiative. There are much fewer measurements of CH_4 and N_2O and it is envisaged that a Global Data

Product for CH_4 and N_2O every 5 years would be sufficient. We have revised the text and Lines 545-557 now read 'The third activity builds on the previous initiative and calls for the production of Global Data Products for dissolved CH_4 and N_2O measurements. To date, individual CH_4 and N_2O measurements are represented at the global scale by the MEMENTO database which has been very successful at compiling CH_4 and N_2O datasets and making them readily accessible to the modeling community. However, the current situation bypasses the important process of compiling a Global Data Product for dissolved CH_4 and N_2O which represents the public release of accumulated quality controlled datasets. The international marine carbon science community has widely embraced such an approach for fCO_2, by submitting data to the Surface Ocean CO_2 Atlas (SOCAT), which was initiated in response to the need for a quality controlled, publicly available, global surface CO_2 dataset (e.g. Bakker et al., 2016). Due to the fewer measurements, a similar data product for marine CH_4 and N_2O would be needed every $\sim$5 years. We consider the production of Global Data Products for dissolved CH_4 and N_2O to be essential for supporting future global modeling efforts and to enhance and reward community engagement'.

Reviewer comment»I would encourage more specific and further steps of practicing these initiatives, such as providing more detailed plans of developing standard operating protocols, preparing reliable reference gases and samples, planning for regular training exercises.

Authors response »We are happy to inform Reviewer #1 that some these activities have been completed while some are still ongoing. A summary table of all these activities is included below for quick reference. The only activity mentioned by Reviewer #1 that is not currently being planned is cross-training exercises due to the ongoing coronavirus pandemic. Ongoing: Standard Operating Protocols (https://web.whoi.edu/methane-workshop/sops/) and production of consensus material for CH_4 and N_2O. Completed: Wilson et al. (2018) Intercomparison exercises; Bullister et al. (2016) Production of compressed gas standards; Kock and Bange (2015) Data portal

Reviewer comment»It is also worthwhile to add some ideas for observations, e.g., episodic/ short-term event monitoring (cyclone disturbance, phytoplankton bloom. . .) and diel rhythm of emission in the coastal zone.

Authors response »Reviewer #1 provides some suggestions here for discrete research projects for CH_4 and N_2O. However, for this overview perspective article, our preference is highlight the availability of analytical tools which can be used to answer any relevant research question and the need for increased coordination among the scientific community. There are multiple examples of this in the text: Application of isotope analysis for methane as mentioned on Lines 241-282 Application of isotopes and isotopomers for nitrous oxide as mentioned on Lines 310-345 Eddy covariance flux towers as mentioned on Lines 499-502 '. . .measurement campaigns in shallow water environments are amenable to the use of eddy covariance flux towers, and they have the potential to lever resources from existing observation networks, which in North America include the Long-Term Ecological Research network (LTER) and the National Estuarine Research Reserve (NERR) System (Novick et al., 2018). Indeed, such activities are already underway; an increasing number of flux towers are being equipped for CH_4 measurements (Torn et al., 2019) and future efforts should focus on the inclusion of N_2O' Development of mobile sampling platforms as mentioned on Lines 426-428 'To determine the contributing factors and resolve the spatial distributions, mobile sampling platforms such as small vessels (Müller et al., 2016; Brase et al., 2017; Tait et al., 2017), and autonomous vehicles (Manning et al., 2019) are essential'.

Reviewer comment»Meanwhile, to better understand and modeling marine CH_4 and N_2O, process study including molecular and isotope approaches from both lab culture and field study could also be added into the database.

Authors response »We interpret this comment by Reviewer #1 to suggest that the MEMENTO data archive for CH_4 and N_2O concentrations could be extended to include other types of datasets including molecular (presumably DNA) and isotopes (presumably natural abundance water-column values). We are strong advocates for data archival in nationally supported databases. These national data repositories lead the way in making datasets publicly available that adhere to FAIR data principles (Wilkinson et al., 2016). With regards to environmental molecular data, all genetic sequence data should be submitted to national databases (e.g., the National Center for Biotechnology Information (NCBI) GenBank database) that provide access to the most up-to-date comprehensive DNA sequence information. Similarly, water-column isotope and concentration datasets should be submitted to national oceanographic data repositories (e.g. BCO-DMO, BODC). The MEMENTO data archive is a specialized collection of CH4 and N2O concentrations, and thereby facilitates the use of these trace gas data by the modeling community, as stated on Lines 172-173: "MEMENTO is now sufficiently mature to support descriptions of the broad-scale surface distributions of CH4 and N2O (e.g. Suntharalingam et al., 2012; Zamora and Oschlies, 2014; Buitenhuis et al., 2018; Battaglia and Joos, 2018)." The datasets should also be deposited in the appropriate national archives to ensure their long-term survival and adherence to FAIR data principles. When submitting data to MEMENTO, there is the option to cross-reference with complementary or co-collected datasets (e.g., DNA or isotope datasets) and also provide a link to publications that include this information. Wilkinson, M.D., Dumontier, M., Aalbersberg, I.J., Appleton, G., Axton, M., Baak, A., Blomberg, N., Boiten, J.W., da Silva Santos, L.B., Bourne, P.E. and Bouwman, J., 2016. The FAIR Guiding Principles for scientific data management and stewardship. Scientific data, 3(1), pp.1-9.

Reviewer comment»In addition to CH4 and N2O observation, the standard measurement of the parameters for air-sea flux calculation, such as the gas transfer velocity or the eddy covariance, should be incorporated to derive accurate air-sea flux.

Authors response »We interpret this comment by Reviewer #1 to suggest that there should be a uniform application of the gas transfer velocity. However, until there is an understanding of which parameterizations are most suitable for the different coastal environments with their inherently different characteristics of fetch, depth, and tidal currents, this is not possible. This is highlighted on Lines 387-397 of the manuscript.

Reviewer comment»Finally, given the profound but unclear impacts of the global change and human activities on the marine carbon and nitrogen cycles, research on CH_4 and N_2O cycling under various external forcing (i.e. deoxygenation, warming, acidification, eutrophication) are encouraged to be incorporated as a component of the database.

Authors response »The manuscript mentioned the influences of different stressors in several places. In the Introduction Lines 103-109 state '. . .the marine environment is susceptible to an accelerating rate of anthropogenic change that will continue to modify the global cycles of carbon and nitrogen into the future. Environmental impacts on marine CH_4 and N_2O distributions include increasing seawater temperatures, decreasing concentrations of dissolved oxygen (O_2), acidification, retreat of ice and mobilization of carbon substrates from former permafrost, altering coastal run-off, and eutrophication (IPCC, 2019)'. In Section 3 on CH_4, Lines 220-2223 state 'Seabed CH_4 emissions are hypothesized to increase in a warming ocean through the decomposition of gas hydrates, the degradation of subsea permafrost under some high-latitude seas, and the increased biodegradation of sediment carbon (Romanovskii et al., 2005; Biastoch et al., 2011; Ruppel and Kessler, 2017; Borges et al., 2019)'. In Section 4 on N_2O, Lines 291-292 state '. . ..make upwelling regions a focal point for N_2O research, particularly since O_2 deficient ocean zones are increasing in size (Stramma et al., 2011)'.

Reviewer comment»The O_2 threshold for denitrification is still controversial, the redox potential is likely to be a better index to explore denitrification and other redox reactions relevant to N_2O and CH_4. In this sense, the measurements of ORP may be included in sampling campaign and database. For modelers, the ORP, which can be connected to electron flow and energy loss-gain, may be useful to advance models with new parameterizations of those chemoautotrophic microorganisms.

Authors response »Reviewer #1 suggests that measuring the oxidation reduction potential (ORP) of a sample is likely to be more informative than O_2 concentrations. ORP measurements are more commonly associated with wastewater and sediments (e.g.

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Tumendelger et al 2019; Zhang et al., 2020) rather than the open ocean for several reasons: (1) Its not only O2 concentrations that are useful but related parameters such as Apparent Oxygen Utilization (AOU) which inform about the deviation from theoretical equilibrium; (2) O2 measurements are nearly always included on every hydrographic CTD cast and it is not evident that commercially available ORP sensors can withstand high pressures. Because of these factors, while we agree with Reviewer #1 that the O2 threshold for denitrification is unresolved, we do not feel that ORP measurements represent a significantly better approach. The manuscript advocates for resolving the relationship between N2O and O2 with increased laboratory based studies. Lines 446-452 state 'For N2O, laboratory studies quantifying microbial process rates, such as for nitrification and denitrification, are relatively few (e.g. Frame and Casciotti 2010; Santoro et al. 2011; Löscher et al. 2012; Ji et al. 2015; Qin et al., 2017). Consequently, models largely continue to use process rates optimized using water column concentrations of N2O, O2, and related nitrogen cycle quantities (e.g. Battaglia and Joos, 2018; Buitenhuis et al., 2018; Landolfi et al., 2017). 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. . .'.. Finally, Reviewer #1 also mentions modeling the flow of electrons but we feel that this is more relevant at the cellular level (e.g. Hink et al 2017) rather than the ecosystem level which is the focus of this manuscript. Tumendelger et al (2019) Methane and nitrous oxide emission from different treatment units of municipal wastewater treatment plants in Southwest Germany. PloS one, 14(1), p.e0209763. Zhang, X., Wang, X., Feng, W., Li, X. and Lu, H., 2020. Investigating COD and Nitrate–Nitrogen Flow and Distribution Variations in the MUCT Process Using ORP as a Control Parameter. ACS omega, 5, 4576-4587. Hink et al (2017) Kinetics of NH3âĂŘoxidation, NOâĂŘturnover, N2OâĂŘproduction and electron flow during oxygen depletion in model bacterial and archaeal ammonia oxidizers, Environ. Microbiol., 19, 4882–4896.

Reviewer comment»The authors synthesized almost all recent documents, which are very useful for beginners who are interested in monitoring marine greenhouse gases.

Overall, this is a well written comprehensive review.

Authors response »Thank you

Reviewer comment»Some problems still, many of their statements or illustrations are not referred specifically to the corresponding figures, for example, Fig. 1a, 1b, 2a, 2b, 2c, 2d and 4. Figure 5a is not mentioned in the text.

Authors response »All figures are now referenced in the text.

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