GENERAL COMMENTS

Currently, there is considerable interest in understanding the production and emissions of greenhouse gases (GHGs) from wastewater treatment plants (WWTPs). Indeed, emissions of these gases have become a major concern in efforts to mitigate climate change and reduce global emissions. This renders the article by Tang et al. particularly significant as it investigates the impact of WWTPs on N_2O emissions in aquatic systems downstream of the Potomac River estuary by measuring nitrogenous nutrients and N_2O concentrations on a monthly resolution over the course of a year. The authors have identified spatially and temporally variable concentrations of N_2O and fluxes of N_2O, generally higher downstream of the WWTPs, highlighting the necessity for effective N_2O removal alongside nitrogen treatment at WWTPs.

The data are well presented and the discussion of the dataset is comprehensive and conclusive. However, from my point of view, I have some suggestions to render the work more attractive to readers. Therefore, I suggest its publication after major revisions.

It would be valuable for the study to clarify whether the three treatment plants (Noman Cole, Mooney, and Aquia) utilize identical wastewater treatment processes and treat similar volumes of water. Additionally, assessing whether the receiving channels into which the WWTPs discharge exhibit comparable water volumes is crucial for ensuring a consistent dilution effect of the gas in the water column. Moreover, understanding the depth of the water column is essential; in cases of shallow depths, the influence of gas emission from the sediment to the water column could be substantial.

It would be interesting for the study to elucidate whether the three treatment plants (Noman Cole, Mooney, and Aquia) employ the same wastewater treatment processes and the volume of water they treat. It is also important to determine if the receiving channels where the WWTPs discharge have similar water volumes, so that the dilution effect of the gas in the water column is similar. Similarly, it would be interesting to know the depth of the water column; if it is shallow, the influence of gas emission from the sediment to the water column could be significant.

The bibliographical references cited do not always follow the same criteria (chronological order or alphabetical order).

SPECIFIC COMMENTS

Ln 49. In a more recent article than those cited, Rosentreter et al., 2023 there are compiled N_2O emissions data from various estuaries, providing a wider range of emissions variation (0.2 – 5.7 Tg N_2O yr^{-1}). Specifically, the paper states: "*Global estimates of estuarine N_2O emissions are highly uncertain, with large discrepancies for both observation-based (220–5,710 $GgN_2O\,yr^{-1}$) and modelling approaches (94–1,084 $GgN_2O\,yr^{-1}$).*

Ln 65-66. References should be listed in ascending chronological order, consistent with the rest of the paper.

Ln 87. It should be indicated what type of treatment is given in the WWTPs (primary, secondary, tertiary, etc.) in order to understand if the nitrogen removal capacity of the three wastewater treatment plants is the same. At what distance from the WWTPs were the samples taken? Were the samples taken at approximately the same distance from the discharge point at all three WWTPs? Were the channels where the samples were taken similar? Did they have approximately the same water volume? An important factor when comparing the amount of N_2O in the receiving channels is dilution.

Ln 99. Were the samples collected from a vessel? Please specify.

Ln 110-11. How was a 3 mL air headspace created in the 60 mL serum bottles? Did all samples have exactly the same volume of air headspace? Could this 3 mL of air in contact with the sample potentially interfere with the measurement? Was the N_2O content in the air also measured? Were the samples taken in duplicate?

Ln 112. Leave a space between the 10 and the M.

Ln 124-128. Figure 4a, depicting the sampling points of the four streams/rivers (Neabsco Creek (5 stations), Occoquan River (3 stations), Pohick Creek (4 stations), and Accotink Creek), should be included in the Materials and Methods section.

Ln 128. Where have the data on water discharge and nitrogen (kg) per day from the wastewater treatment plants been obtained? It would be interesting to include this information in the manuscript.

Ln 149. It is not reflected in the text how the 3 mL air headspace is taken from the serum bottles to estimate the amount of N_2O in the sample.

Ln 167. *"The equilibrium N_2O concentration was calculated based on the solubility of N2O (Weiss and Price, 1980)…"* Where did you obtain the value of N_2O in the atmosphere for the calculations? Which value did you consider, the daily, monthly...?

Ln 170: What do the initials NCEP stand for? It would be more comprehensive to include the website from which the value of U was taken.

Ln 171. You should cite in the paper the expression from which Sc has been estimated, possibly from the proposed expression by Wanninkhof (2014). You should indicate whether for the calculation of Sc, you have considered the expression for salinity equal to zero, or if, on the contrary, the N_2O Schmidt number for each point has been scaled to the values proposed by Wanninkhof (2014) for salinities between 0 and 35, assuming that Sc varies linearly with salinity.

Ln 173. References should be listed in ascending chronological order, consistent with the rest of the paper.

Ln 170-176. I don't understand why they are using a gas transfer velocity parameterization (k) proposed for the ocean, such as the expression by Wanninkhof (2014), rather than a k for a coastal system. If they didn't have data on current velocity and depth of the system necessary to use the k by Borges et al. (2004) and Rosentreter et al. (2021), they could have used the expression proposed by Raymond and Cole (2001), which is based on a compilation of k proposed for different coastal systems, or that by Jiang et al. (2008), based on the compilation of Raymond and Cole (2001) as well as other studies conducted in estuaries. Furthermore, given the uncertainty associated with k, to minimize this, they could have estimated water-atmosphere fluxes considering two expressions of k (Raymond and Cole, 2001; Jiang et al., 2008; Wanninkhof, 2014), and taken the average value of the three fluxes obtained, as many other authors do in coastal systems (e.g., Call et al., 2015; Sánchez-Rodriguez et al., 2024)

Ln 233. I suggest wording it like this: ….vs 6‰ for stations of the central channel and without the influence by WWTPs

Ln 242. In general, denitrification typically occurs in environments with low oxygen concentrations (DO ≤ 5 μmol L−1, Codispoti et al., 2001). As illustrated in Supplementary Figure 2, oxygen concentrations at the stations never reached low values. In fact, downstream stations of wastewater treatment plants exhibited dissolved oxygen levels ranging between 139.38 (25/07/2022) – 430.94 μM (7/02/2023). It is recognized that denitrification can also take place within oxygenated water columns containing suspended organic matter particles (Bange, 2008). Is there a substantial amount of suspended material in the studied system that could induce denitrification in oxygenated water? On the other hand, it is well-established that coastal sediments provide optimal environments for denitrification due to continuous inputs of nutrients and organic matter from land. Could it be that some of the measured N_2O in the water originates from the sediment?

Ln 252. Correlations of 0.62 (r^2=0.38) and 0.51 (r^2=0.26), I do not consider them strong correlations, remove the word strong.

Ln 252-256 and 263-264. In stations unaffected by WWTPs, there appears to be a good positive correlation between N_2O and DO and NOx, which could indicate that nitrification is an important process in these N_2O production stations.

Ln 262. References should be listed in ascending chronological order, consistent with the rest of the paper.

Ln 278-279. References should be listed in ascending chronological order, consistent with the rest of the paper.

Ln 292-294. Why does it not also present the predictive model of N_2O concentration based on total nitrogen and temperature for stations in the central channel of the Potomac Estuary? It could be interesting to have it to extrapolate to other areas of the estuary located in the channel. Perhaps you have included the data measured in the channel in the

samples without wastewater treatment plants (WWTPs). If so, please indicate it. I believe you should have stated the number of stations/data considered in each prediction.

Ln 298-300. Did you use the prediction model for stations without WWTPs? Please indicate it in the paper.

FIGURES

Figure 2 and Supplementary Figure 1. What does the "01" after the slash indicate on the x-axis of the central graphs? Wouldn't it be more intuitive for the reader to use "22" or "23" instead of "01," depending on the year the sampling was conducted?

Supplementary Figure 2. In the figure caption and in Figure a, a negative sign as a subscript is missing on NOx^- on the x-axis. In Figure b, on the x-axis, remove the space between N_2 and O.

REFERENCES:

Bange, H. W., 2008. Gaseous Nitrogen Compounds (NO, N_2O, N_2, NH_3) in the Ocean. In Nitrogen in the Marine Environment (pp. 51–94). Elsevier Inc. https://doi.org/10.1016/B978-0-12-372522-6.00002-5

Call, M., Maher, D. T., Santos, I. R., Ruiz-Halpern, S., Mangion, P., Sanders, C. J., ... & Eyre, B. D., 2015. Spatial and temporal variability of carbon dioxide and methane fluxes over semi-diurnal and spring–neap–spring timescales in a mangrove creek. Geochimica et Cosmochimica Acta, 150, 211-225.

Codispoti, L.A., Brandes, J.A., Christensen, J.P., Devol, A.H., Naqvi, S.W.A., Paerl, H.W., Yoshinari, T., 2001. The oceanic fixed nitrogen and nitrous oxide budgets: moving targets as we enter the anthropocene? Sci. Mar. 65 (S2), 85–105.

Jiang, L.Q., Cai, W.J., & Wang, Y. (2008). A comparative study of carbon dioxide degassing in river- and marine-dominated estuaries. Limnology and Oceanography, 53(6), 2603–2615.

Rosentreter, J. A., Laruelle, G. G., Bange, H. W., Bianchi, T. S., Busecke, J. J. M., Cai, W. J., Eyre, B. D., Forbrich, I., Kwon, E. Y., Maavara, T., Moosdorf, N., Najjar, R. G., Sarma, V. V. S. S., Van Dam, B., & Regnier, P., 2023. Coastal vegetation and estuaries are collectively a greenhouse gas sink. Nature Climate Change, 13(6), 579–587. https://doi.org/10.1038/s41558-023-01682-9

Sánchez-Rodríguez, J., Ortega, T., Sierra, A., Mestre, M., Ponce, R., Fernández-Puga, M. D. C., & Forja, J., 2024. Distribution, reactivity and vertical fluxes of methane in the Guadalquivir Estuary (SW Spain). Science of The Total Environment, 907, 167758.