Responses to reviewers' comments on "Global Nitrous Oxide Budget 1980–2020" (manuscript number essd-2023-401)

We would like to thank the reviewers for their thoughtful and insightful comments. The manuscript has been revised accordingly, and our point-by-point responses in blue color are provided below, and our new/modified texts in the revised manuscript are indicated in red color.

Reviewer 3:

This paper is an ambitious and detailed compilation of information across many data sources and different modeling approaches. As such, it will provide a useful reference for the scientific community, including policy makers.

Response: We thank the reviewer for the positive comments!

I have listed my most major comments below.
The paper does not always clearly distinguish between results that are highly speculative and purely model based (e.g., responses to CO2 and climate change) and those that have a more solid grounding in data (e.g., responses to fertilizer and manure inputs). For example, lines 123-131 are stated as though they are facts. They should be qualified with, "according to BU estimates," as is appropriately done for the presentation of model results in the next paragraph starting on line 132.

Response: Thank you for your insightful suggestion! We acknowledge the importance of distinctly differentiating between speculative, model-based results and those grounded in data. We have added "According to BU estimates," to the revised manuscript.

It also would be useful to include a graphical depiction of the relative uncertainty of different budget terms in Figure 1, e.g., perhaps with solid arrows for more robustly known fluxes and more faded arrows for speculative fluxes. (The current arrows have more faded colors that transition to more solid colors but it is not clear what this transition represents.)

Response: Thanks for the suggestion. We have tried to implement it, but the final result was not satisfactory from a visual point of view. We hope that the fact each flux value comes with a clear range, as an indication of its uncertainty, provides all the information necessary for the reader to know that not all fluxes are known the same.

Line 217 "Reducing N2O emissions is a required net-zero greenhouse gas (GHG) emission and the recovery of stratospheric ozone." First, this sentence does not make sense grammatically. Second and more importantly, the concept of "net zero" is not appropriate for N2O. Earth has always been, and always will be, a net natural source of N2O, which is then destroyed photochemically in the stratosphere. "Net zero" is mentioned again on line 298 so line 217 does not seem to be just a typo. I bring this up because I have seen essays arguing for "N2O neutrality" as a feasible and desirable policy goal, to the point that spraying chemicals on natural landscapes is considered as a way to stop nitrification. The idea that humans should be actively trying to stop natural N2O emissions seems likely to have potentially bad unintended

consequences. The concept that "net zero" is not logical for N2O (which is not analogous to CO2) should be clearly communicated to policy makers, who may not be familiar with Earth's natural biogeochemical cycles.

Response: Thanks for pointing this out! We agree with the reviewer on that Earth surface is a net natural source of N_2O and N_2O neutrality is not a feasible and desirable policy goal. We have revised the sentences to avoid misleading readers and policymakers:

"Reducing N_2O emissions will contribute to the mitigation of global warming and the recovery of stratospheric ozone (Jackson et al., 2019)."

"contribute to the global stocktake of the Paris Agreement to track progress towards national determined contributions."

There is a confusing switching back and forth between 3 alternative time frames: 1997-2020, 1980-2020 and 2010-2019. Please state clearly somewhere early in the methods why these 3 time frames were chosen and why each is significant.

Response: "We focus on N_2O fluxes and their change rates during three periods: 1997-2020, 1980-2020 and 2010-2019. 1980-2020 is the entire study period, we report temporal variations in BU estimates of N_2O emissions from different sources to depict the overall trends of these fluxes. 1997-2020 is the overlapping period of BU and TD approaches, we compare BU and TD estimates during this period to exam their consistency. 2010-2019 is the most recent decade, we report the magnitudes of emissions from different sources to show their latest status and relative importance."
We have added these statements to the revised manuscript.

The frequent reporting of rates of increase in TgN/yr-2 units is not intuitively meaningful and arguably not mathematically correct. Strictly speaking, it assumes that the source can be fit with a parabolic (t^2) parabolic dependence on time, which is not obviously the case for many regions, based on figure 14. At minimum please explain how these rates of rates of increase were calculated and why there is so much emphasis on reporting them.

Response: We are sorry for the unclear statement. As stated in Section 2.1, annual N_2O fluxes are expressed in teragrams of N_2O-N per year: Tg N_2O-N yr^{-1} (Tg N yr^{-1}), and the change rates in N_2O fluxes are expressed in the unit of Tg N_2O-N yr^{-2} (Tg N yr^{-2}). Therefore, rates of increase or decrease reported in this paper are the first derivatives of annual N_2O fluxes, rather than the second derivatives. They represent the average change rate
We have revised the sentence to avoid confusion:

"In this study, N_2O fluxes are expressed in teragrams of N_2O-N per year: 1 Tg N_2O-N yr^{-1} (1 Tg N yr^{-1}) $=10^{12}$ g N_2O-N yr^{-1}=1.57×10^{12} g N_2O yr^{-1}, with change rates in N_2O fluxes expressed in the unit of Tg N_2O-N yr^{-2} (Tg N yr^{-2}) which represent the first derivative of annual N_2O fluxes calculated by the linear regression method."

Below is a list of more detailed minor comments.

Line 84. It would be useful to provide an estimate of N2O's relative contribution to enhanced GH forcing (e.g., 6% or whatever the latest value is).

Response: Thanks for the suggestion. We have added the following sentence into the revised manuscript: "According to Forster et al (2023), N_2O's relative contribution to the total enhanced effective radiative forcing of greenhouse gases was 6.4% for 1750-2022."

Line 109. It's odd to mention 10% here when the abstract cited "nearly 25%" from the preindustrial, indicating that 15% of the rise was prior to 1980. This isn't wrong necessarily but it sends a confusing message.

Response: Thanks for the suggestion! We have revised this sentence according to your suggestion:
"The tropospheric N_2O mole fractions, precisely measured at a global network of stations, increased from 301 parts per billion (ppb) in 1980 to 333 ppb in 2020 and 336 ppb in 2022."

Line 110. What does "it" refer to?

Response: We are sorry for the unclear statement. "It" refers to the tropospheric N_2O mole fraction in 2022. We have revised this sentence:

"The tropospheric N_2O mole fraction in 2022 is higher than at any time in the last 800,000 years."

Line 113. I suggest to delete "with a substantially lower resolution" because it is confusing.

Response: Thanks for the suggestion, and we have deleted it according to your suggestion.

Lines 123-131. This paragraph should state explicitly that these results (i.e., trends in different sources) are based on BU approaches, since TD approaches cannot generally distinguish individual sources. As such, the BU results are in large part based on speculative model-based estimates. The paragraph states them as though they are known facts.

Response: Thanks for your insightful suggestion! We have added "According to BU estimates," in the manuscript.

Lines 132-144. This paragraph seems to overlap considerably with the previous paragraph (lines 117-131). Is the previous paragraph covering the period 1997-2020 (or 1980-2020?), while the current paragraph covers 2010-2019? Please distinguish the 2 paragraphs more clearly and consider deleting one of them unless there is a compelling reason to distinguish the last 10 years from the last 23-40 years.

Response: Line 117-131 focuses on reporting the temporal variations in N_2O emissions over 1980-2020 (the entire study period) and compare the trends in BU and TD estimates during their overlapping period (1997-2020). Line 132-144 shows the

magnitudes of emissions from different sources and their relative importance in the most recent decade. Moreover, line 117-131 focuses on the total emissions or emissions from the five big categories, while line 132-144 also reports emissions from the 21 identified natural and anthropogenic sources which contains more detailed information. In summary, these two paragraphs have different emphases. Therefore, we think it is better to keep both paragraphs.

Line 149. "since" is confusing. Do you mean "by"?

Response: Yes! We have corrected it.

Line 151. Is this a second derivative (Tg N/yr-2)? I think this will be confusing to many readers and not intuitively meaningful. I suggest to present as the rate of growth in TgN/yr in the 1980s contrasted with the higher rate of growth in the most recent decade.

Response: We are sorry for the unclear statement. As stated in Section 2.1, N_2O fluxes are expressed in teragrams of N_2O-N per year: Tg N_2O-N yr^{-1} (Tg N yr^{-1}). Therefore, it is the first derivative of annual N_2O fluxes, rather than the second derivative. We have revised the sentence to avoid confusion:

"In this study, N_2O fluxes are expressed in teragrams of N_2O-N per year: 1 Tg N_2O-N yr^{-1} (1 Tg N yr^{-1}) =10^{12} g N_2O-N yr^{-1}=1.57×10^{12} g N_2O yr^{-1}, with change rates in N_2O fluxes expressed in the unit of Tg N_2O-N yr^{-2} (Tg N yr^{-2}) which represent the first derivative of annual N_2O fluxes calculated by the linear regression method."

Line 169. What is "manure forest conversion"?

Response: We are sorry for the spelling error. It should be "mature forest conversion". We have corrected it in the manuscript.

Line 198. 66/270 = 24.4%, which is not "more than 25%".

Response: We are sorry for the calculation error. We have corrected it in the manuscript.

"Atmospheric N_2O mole fractions have increased by more than 24% since the pre-industrial era"

Line 203. Please update the NOAA reference.
Lan, X., E.J. Dlugokencky, J.W. Mund, A.M. Crotwell, M.J. Crotwell, E. Moglia, M. Madronich, D. Neff and K.W. Thoning (2022). Atmospheric Nitrous Oxide Dry Air Mole Fractions from the NOAA GML Carbon Cycle Cooperative Global Air Sampling Network, 1997-2021, Version: 2022-11-21, *https://doi.org/10.15138/53g1-x417*.

Response: Thank you! We have updated the NOAA reference.

Line 207. Please clarify that this means since the period of observations began. As written, it seems to imply that the growth rate was higher prior to 1980.

Response: Yes, this means since the period of observations began. We have deleted "since 1980" to avoid confusion.

Line 217. What does "is a required net-zero GHG emission" mean?
Response: We are sorry for the grammatical error. We have revised the sentences as follows:

"Reducing N_2O emissions will contribute to the mitigation of global warming and the recovery of stratospheric ozone (Jackson et al., 2019)."

Line 223. This suggests that up to 44% of N2O is produced abiotically. This doesn't seem right. Furthermore the cited 56-70% seems at odds with Figure 1, in which only 2.3/18.1 TgN (12.7%) is from a non-microbial source.

Response: Thank you for the suggestion. This sentence has been revised:
"Nitrification and denitrification are the two key microbial processes controlling N_2O production, making the largest contribution to global N_2O emissions;"

Line 248. Grammar note: missing "a" before small.

Response: Thanks for pointing out this grammar error. We have revised accordingly.

Substance note: the effect on the N2O source from OMZs in the ocean will not necessarily be small, but perhaps this sentence refers to the fact that N2O contributes a relatively small part of GHG warming (even if the expanding OMZ effect on N2O emissions is large).

Response: Yes, we acknowledge that the effect on the N_2O emissions from OMZs in the ocean may be large. This sentence refers to the fact that ocean N_2O emission contributes a relatively small part of GHG warming.

Figure 3. Please delete page number 275.

Response: We have deleted it.

Line 298. Again, it is not possible or even desirable to achieve net zero emissions of N2O. To do so would involve severe disruptions to Earth's natural nitrogen cycle. This needs to be made clear to policy makers who are not familiar with biogeochemistry.

Response: Thanks for pointing this out! We agree with the reviewer on that Earth surface is a net natural source of N_2O and N_2O neutrality is not a feasible and desirable policy goal. We have deleted "and the ultimate goal of achieving net-zero GHG emissions" to avoid misleading readers and policymakers.

Line 355. It is debatable whether models are accurately capturing nitrification, denitrification, and other key processes (e.g., Nevison, C., Goodale, C., P. Hess, W.R. Wieder, J. Vira and P.M. Groffman (2022). Nitrification, and denitrification in the Community Land Model compared to observations at Hubbard Brook Forest. Ecological Applications. https://doi.org/10.1002/eap.2530.)

Response: Thanks for pointing this out! We acknowledge that it is debatable whether process-based models can accurately capture nitrification, denitrification, and other key processes. We revised the sentence as follows:

"they are capable of modelling the key processes affecting N_2O production and emission such as autotrophic nitrification, denitrification, plant nitrogen uptake, ammonia volatilization, nitrate leaching, soil thermal and hydrological processes, although their accuracy in representing these processes needs further improvement;"

p.15, Table 1. Please specify whether the shelf products represent natural or anthropogenic emissions or both.

Response: We categorized shelf emissions into the natural emissions category. We have specified it in the Table 1.

p.16, Table 1. Please indicate what type of emissions are modeled with these "other" approaches.

Response: Thank you for the suggestion! We have added the type of emission modeled by SRNM, bookkeeping method, and IMAGE-GNM in Table 1.

Line 419. That this part of the natural flux was ASSUMED to be constant should be acknowledged in the abstract around line 130, which states that natural sources were relatively constant, as though this were a result.

Response: Thank you for this suggestion. Among all sources, natural emissions from shelves, inland waters, and lightning and atmospheric production were assumed to be constant during 1980-2020. We have added this sentence to the manuscript:

"Among all sources, natural emissions from shelves, inland waters, and lightning and atmospheric production were assumed to be constant during 1980-2020. According to BU approaches, the total natural emissions from these sources were 1.7 (0.9-3.0) Tg N yr^{-1}."

Line 423. 44% of what?

Response: 44% of the total N_2O emissions from inland waters. We are sorry for the unclear statement. The results in Yao et al. (2020) suggested that 56% of the total N_2O emissions from rivers, reservoirs, estuaries and lakes were attributed to anthropogenic N additions, and the resting 44% of the total N_2O emissions were from natural sources. To avoid confusion, this sentence has been revised as:

"Using this approach, we estimated that N_2O emissions from natural sources of rivers, reservoirs, lakes and estuaries accounted for 44% (36%−52%) of the total emissions from inland waters."

Line 492. Please comment here on whether any data exist to evaluate these model predictions. This seems like a highly speculative flux to include in the budget, with a less solid grounding in data and observations than, e.g., direct agricultural emissions from fertilizer or manure.

Response: We acknowledge that N_2O fluxes from climate/CO_2/land cover change are highly uncertain. Although data from control experiments exist at several sites, no global or regional level data exists to evaluate model predictions.

Line 505. Is this in the stratosphere or the troposphere?

Response: This is in the stratosphere. We are sorry for the unclear statement. We have revised the sentence:

"There is also N_2O production from N2 +O(1D), which amounts to about 2% of the atmospheric source in the stratosphere (Estupiñán et al. 2002)."

Line 587. Why such a small increase in manure management when the other 3 sources increased by 50% or more during the same period?

Response: Emissions from agriculture-related activities in EDGAR and FAOSTAT, including direct and indirect N_2O emissions and manure management are estimated based on the IPCC methodology. IPCC coefficients for manure management are skewed towards developed countries, where animal numbers have stayed rather constant overall—and diminished in Europe for instance. Conversely, manure left on pasture is applied to all countries and hence heavily influenced by increasing animal trends in the rest of the world. For manure management sector, the technology penetration in some countries might offset the increases in emissions associated to the increases in number of heads.

Line 626. What is EDGAR/NMIP2? I do not see it described above in the methods.

Response: Sorry, it should be NMIP2/EDGAR v7.0, we have corrected it. We define "NMIP2/EDGAR v7.0" in section 2.4.4: "EDGAR v7.0 provided estimates of indirect emissions from both agricultural and non-agricultural sectors, however, here, we sum the ensemble mean of NMIP2 estimates of indirect emissions from agricultural sectors with indirect emissions from non-agricultural sector of EDGAR v7.0 (i.e., NMIP2/EDGAR v7.0) to represent N deposition induced soil emissions from both agricultural and non-agricultural sectors."

Line 700. Perhaps comment on whether the trend in the posterior fluxes differed substantially from the assumed trend in the prior emissions.

Response: Thanks for the suggestion. We have revised accordingly.

Figure 13. Please label TD and BU in the figure legend and/or describe in the caption.

Response: We are sorry for the unclear statement. We have added more statements in the figure caption:

"The blue lines represent the mean N_2O emission from bottom-up methods and the shaded areas show minimum and maximum estimates; the red lines represent the mean N_2O emission from top-down methods and the shaded areas show minimum and maximum estimates."

Line 798. Similar to my comment in the executive summary, why are 1997-2020 and 1980-2020 used as alternative historical periods?

Response: 1980-2020 is the entire study period, we report temporal variations in BU estimates of N_2O emissions from different sources to depict the overall trends of these fluxes. 1997-2020 is the overlapping period of BU and TD approaches, we compare BU and TD estimates during this period to exam their consistency. We have added these statements to the revised manuscript.

Line 898. Was this decrease due mainly to a reduction in fertilizer use from 1980 to 2020? If so, readers might be interested to know if the reduction was this achieved due to deliberate mitigation strategies or rather to the collapse of the Soviet Union?

Response: This decrease was mainly caused by a reduction in fertilizer use after the collapse of the Soviet Union (Tian et al., 2022). We have revised this sentence:

"Direct agricultural emissions and indirect emissions show overall decrease trends from 0.46 and 0.16 Tg N yr^{-1} in 1980 to 0.38 and 0.12 Tg N yr^{-1} in 2020, respectively, mainly due to a reduction in fertilizer use after the collapse of the Soviet Union (Tian et al., 2022)."

Line 998 and elsewhere. Again, the use of the second derivative is confusing.

Response: We are sorry for the unclear statement. They are first derivatives of annual N_2O fluxes, rather than second derivatives. We have revised the description of units of N_2O fluxes to avoid confusion:

"In this study, N_2O fluxes are expressed in teragrams of N_2O-N per year: 1 Tg N_2O-N yr^{-1} (1 Tg N yr^{-1}) =10^{12} g N_2O-N yr^{-1}=1.57×10^{12} g N_2O yr^{-1}, with change rates in N_2O fluxes expressed in the unit of Tg N_2O-N yr^{-2} (Tg N yr^{-2}) which represent the first derivative of annual N_2O fluxes calculated by the linear regression method."

Line 1112. Again, what is manure forest conversion?

Response: We are sorry for the spelling error. It should be "mature forest conversion". We have corrected it in the manuscript.

References:

Estupinán, E. G., et al. "Investigation of N_2O production from 266 and 532 nm laser flash photolysis of $O_3/N_2/O_2$ mixtures." *The Journal of Physical Chemistry A* 106.24 (2002): 5880-5890.

Forster, Piers M., et al. "Indicators of Global Climate Change 2022: annual update of large-scale indicators of the state of the climate system and human influence." *Earth System Science Data* 15.6 (2023): 2295-2327.

Tian, H., et al. "History of anthropogenic Nitrogen inputs (HaNi) to the terrestrial biosphere: a 5 arcmin resolution annual dataset from 1860 to 2019." *Earth System*

Science Data 14.10 (2022): 4551-4568.

Yao, Y., et al. "Increased global nitrous oxide emissions from streams and rivers in the Anthropocene." *Nature Climate Change* 10.2 (2020): 138-142.