

point-by-point response to the reviews.

Response to the Reviewer #1

The authors have responded carefully and thoroughly to the original reviews. I have just a few more comments, focused on the beginning sections of the manuscript.

Response: We would like to thank the reviewer for the careful review and positive 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**.

Line 99. Should “biochemical” be “biogeochemical”?

Response: Thanks for the suggestion, we have revised accordingly.

Lines 112-114. “The observed atmospheric N₂O concentrations in recent years have exceeded projected levels under all scenarios in the Coupled Model Intercomparison Project Phase 6 (CMIP6), underscoring the urgency to reduce anthropogenic N₂O emissions.” The word “urgency” seems overstated, given that N₂O contributes a relatively small 6.4% of anthropogenic radiative forcing, as stated above on line 96. I recommend changing “urgency to reduce” to something like “importance of reducing.” There could be many reasons why CMIP6 underestimated the rate of growth. It is therefore perhaps not appropriate to create alarm on that basis, especially without context or explanation of how far in excess the actual concentration is compared to the CMIP6 projected concentration.

Response: Thanks for the suggestion, we have revised accordingly.

Line 121-122. There are some missing words in this sentence, which I have inserted as capitals, “Ice core data show A relatively constant tropospheric N₂O mixing ratio over the past two millennia (Canadell et al., 2021; MacFarling Meure et al., 2006; Fischer et al., 2019), FOLLOWED BY AN INCREASE from about 270 ppb in 1750 to well above 300 ppb.”

Response: Thanks for the suggestion, we have revised accordingly.

Line 133. “BU approaches estimated...” would be better than “BU approaches showed...”

Response: Thanks for the suggestion, we have revised accordingly.

Lines 237-238. “Reducing N₂O emissions will contribute to the mitigation of global warming and the recovery of stratospheric ozone (Jackson et al., 2019).” This would be a good place to include the important note that stratospheric NO_x from N₂O has offset (rather than exacerbated) halogen-catalyzed stratospheric ozone loss through various buffering reactions, e.g., the formation of halogen reservoir species like ClONO₂. NO_x also buffers HO_x-catalyzed ozone loss. As a result, increased stratospheric NO_x due to rising levels of N₂O can lead to incremental

stratospheric O₃ loss but is unlikely to cause catastrophic ozone loss the way that anthropogenic halogens did (e.g., Wennberg et al., 1994; Nevison et al., 1999; Ravishankara et al., 2009).

Response: We have added one sentence according to your suggestion, line 239-243: It is noted that although increased stratospheric NO_x due to rising levels of N₂O can lead to incremental stratospheric O₃ loss but is unlikely to cause catastrophic ozone loss the way that anthropogenic halogens did, because stratospheric NO_x from N₂O has offset halogen-catalyzed stratospheric ozone loss through various buffering reactions, e.g., the formation of halogen reservoir species like ClONO₂ (Wennberg et al., 1994; Nevison et al., 1999; Ravishankara et al., 2009).

Lines 238-241 “Significant reductions of N₂O emissions are required along with net CO₂-emissions to stabilize the global climate.”

This sentence is much improved over the original. However, the subsequent sentence seems to make the debatable assumption that all GHGs should be cut proportionally. This is not necessarily true if some GHGs (e.g., methane) are easier and less societally disruptive to cut than others or if they are shorter-lived, such that their reduction will more effectively reduce radiative forcing. Since anthropogenic N₂O emissions are fundamentally related to food production and the livelihood of farmers, I would encourage a carefully worded statement here. One suggestion would be to add a qualifying clause such as, “For pathways consistent with the remaining carbon budget of 1.5°C, 1.7°C and 2°C stabilization, and assuming that all GHGs should be cut in equal proportion to their contribution to anthropogenic radiative forcing, global N₂O emissions need to be reduced by 22%, 18% and 11 %, respectively*, by 2050 (Rogelj and Lamboll, 2024). *Note that respectively was misspelled.

Response: Thank you! We have added “and assuming that all GHGs should be cut in equal proportion to their contribution to anthropogenic radiative forcing,” to line 245-246 according to your suggestion.

Line 361. The 3 different time frames, which were quite confusing in the original manuscript, are better explained with the addition of this paragraph. However, the choice of 2010-2019 is still confusing since the full period of the study is 1980-2020. It would seem more logical to use 2011-2020 as the last 10 years. I would suggest switching from 2010-2019 to 2011-2020 if this is not too difficult to do. However, this change is not absolutely necessary if it is too burdensome to implement.

Response: Thanks for the suggestion! We chose 2010-2019 because our study is designed to report N₂O budget for the four decades: 1980s, 1990s, 2000s, and 2010s, and we use the year 2020 to represent the most recent status. Therefore, we prefer keeping the choice of 2010-2019, rather than changing to 2011-2020.

References

Wennberg, P.O., et al., 1994. Removal of stratospheric O₃ by radicals: In situ measurements of OH, HO₂, NO, NO₂, ClO and BrO, *Science*, 266, 398-404.

Nevison, C.D, S. Solomon and R.S. Gao, 1999. Buffering interactions in the modeled response of stratospheric O₃ to increased NO_x and HO_x, *J. Geophys. Res.*, 104 (D3), 3741-3754.

Ravishankara, A., Daniel, J. & Portmann, R. 2009. The dominant ozone-depleting substance emitted in the 21st century. *Science* 326, 123-125.

Response to the Reviewer #2

We would like to thank the reviewer for the careful review 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](#).

Direct N₂O “precisely measured at a global network of stations”. Uncertainties known and specified by those sources (e.g CSIRO, NOAA); they spend lots of effort to ensure precision. Not reported here! Would affect e.g. Figure 5!

[Response: We acknowledge that there are uncertainties in measurements from the observation networks \(AGAGE, NOAA and CSIRO\), and we have reported the uncertainties in Figure 2. We didn't plot all the three measurements in Figure 5 because the differences in the three lines are hard to distinguish and the uncertainty information has already been reported in Figure 2.](#)

This reader accepts authors' claims of massive unique effort to gather and report N₂O information, to “explore the relative temporal and spatial importance of multiple sources and sinks”. Not their job, they correctly claim, to construct an encompassing end-to-end uncertainty budget. But, if this group of experts can not or will not undertake such an effort, who will? My review concedes the effort required but remains dismayed by large unspecified unquantified uncertainties. My challenge / hypothesis stands: “uncertainties remain so large so as to preclude conclusions on specific sources or assigned to specific regions”. Again, who, if not this group, will help us sort such issues?

[Response: Thanks for your comments! We acknowledge that our current N₂O budget still has large uncertainties although we have used a large range of available sources to provide an overall assessment. However, our study identifies where the uncertainties are well constrained or very large, which can be viewed as a success of our effort. We also acknowledge that constructing an encompassing end-to-end uncertainty budget will guide future research. We will make more efforts to attempt an improved quantitative analysis of uncertainties in the next round of the N₂O budget.](#)

In opening paragraphs, for example, authors switch between ppm without plus/minus to decadal emissions rates (likewise without min/max designation) to BU source information with min/max. Set aside weaknesses of min/max; why do readers encounter them in some places but miss them completely in others?

[Response: We did not use the unit “ppm” in our study. We used plus/minus to report the magnitude of uncertainties in atmospheric burden in Section 2.5.1. Here, our aim was to report uncertainty itself, so we didn't use the mean \(minimum-maximum\) as in other places. We think it is clear for readers, and will not cause misunderstanding.](#)

Format and labels of figures remain incomplete. Abbreviations applied in Fig 3, for example, remain inconsistent with text and incomplete.

Response: Sorry for the errors. We have added abbreviations of the identified N₂O sources and sinks shown in the main text and corrected the inconsistent descriptions. We also revised Figure 3.

Line 358 delineates data into “three” periods: “1997-2020, 1980-2020 and 2010-2019.” But, at line 320, reader learned that “budgets cover the decades of 1980-89, 1990-99, 2000-09, 2010-2019,”. What?

Response: Our study period is 1980-2020, the statement “budgets cover the decades of 1980-89, 1990-99, 2000-09, 2010-2019 ..” is not contradictory with the statement that “We focus on N₂O fluxes and their change rates during three periods: 1997-2020, 1980-2020 and 2010-2019”.

Line 362: authors claim to “show”; this reader accepts only that they have made best estimates. Term ‘estimate’ frequently used in following paragraphs. A bit of modesty in describing the outcome could have perverse effect of highlighting massive efforts!

Response: Thanks for the suggestion! We have revised the sentence as follows: “we report the magnitudes of emissions from different sources to give best estimates of their latest status and relative importance.”

Line 1323: something wrong here as EDGAR min and max for N emissions due to energy both equate to 113.3%? Or, next line, to 111.8% and 111.8%. Not my job to chase down this reference! Authors need to ensure correct extraction and citation!!! 300% errors? Sentence following, assigning EDGAR as “very uncertain” seems redundant as well as erroneous? EDGAR folks would disagree? All due to erroneous EFs? No means to estimate reliability of national or regional or global EFs? Fig 18 shows EFs ranging from <1 to >3 for productive agricultural regions across the globe. This reader maintains that Fig 18 invalidates prior effort(s) at regional assignments. Likewise for Figs 19 & 21?

Response: The emission factor shown in Figure 18 is derived from NMIP2 models whose estimates were integrated in this study. This figure reflects the phenomenon that emission factor is spatially heterogeneous, which is reported in previous studies (Cui et al., 2021; Harris et al., 2022). There is no direct linkage between spatial heterogeneity of emission factor and the reliability of estimates of emissions, therefore, we don’t agree with the opinion that Fig 18 invalidates our prior effort(s) at regional assignments. Figures 19 and 21 show the uncertainties in estimates of N₂O emissions from soils and ocean, identifying where the uncertainties are well constrained or very large. It can be viewed as a success of our effort, which will inform future research.

This manuscript doesn't implement additional uncertainty analysis for EDGAR database. These uncertainty estimates are directly taken from Solazzo et al. (2021). We modified the statements as follows: "The uncertainties for EDGAR N₂O emissions estimated by Solazzo et al. (2021) are based primarily on the uncertainties in emissions factors and activity data statistics from the IPCC (2006). Globally, these emissions are accurate within an interval of ±113 for energy, -12% to +16% for industrial processes and product use, -225 to +302 for agriculture, -159% to 203% for waste and ±112% for others; the most uncertain emissions are those related to N₂O from waste and agriculture."

Supplement includes only min-max? (If additional info on statistical uncertainties remains buried in individual descriptions of biogeography models or atmospheric inversions this reviewer did not find them.) I want to credit authors for adding uncertainty info, but because I did not look at prior Supplement I can not tell.

Response: In supplementary material, we listed atmospheric N₂O dry mole fraction measured by the three observing networks, and the corresponding minimum and maximum values, as well as minimum and maximum estimates of future projections of atmospheric N₂O dry mole fraction. We reported the minimum and maximum values as an indication of the uncertainty, consistent with the main text.

I admire massive effort to assemble N₂O info from so many disparate sources. If efforts to delineate composite uncertainties have not kept up, that remains mostly a challenge to future research.

Response: Thanks for your insightful comments! We acknowledge that delineating composite uncertainties in N₂O budget will guide future research. We will make more efforts on an improved quantitative analysis of uncertainties in the next round of the N₂O budget.

Reference:

Cui X, Zhou F, Ciais P, et al. Global mapping of crop-specific emission factors highlights hotspots of nitrous oxide mitigation[J]. Nature Food, 2021, 2(11): 886-893.

Harris E, Yu L, Wang Y P, et al. Warming and redistribution of nitrogen inputs drive an increase in terrestrial nitrous oxide emission factor[J]. Nature Communications, 2022, 13(1): 4310.