Response to Editors and Reviewers

We gratefully thank the reviewers for the constructive comments and suggestions to improve the manuscript. As detailed below, the reviewers' comments are shown in *black italic*; our response to the comments is in blue. New or modified text is in red.

Responses to Referee 1:

Updated Figure 1 still has solid lines for all regions. Please check all the figures again.

Response: Thanks for your feedback. We have corrected the solid line issue in Figure 1 and have thoroughly checked and revised all figures in the manuscript.

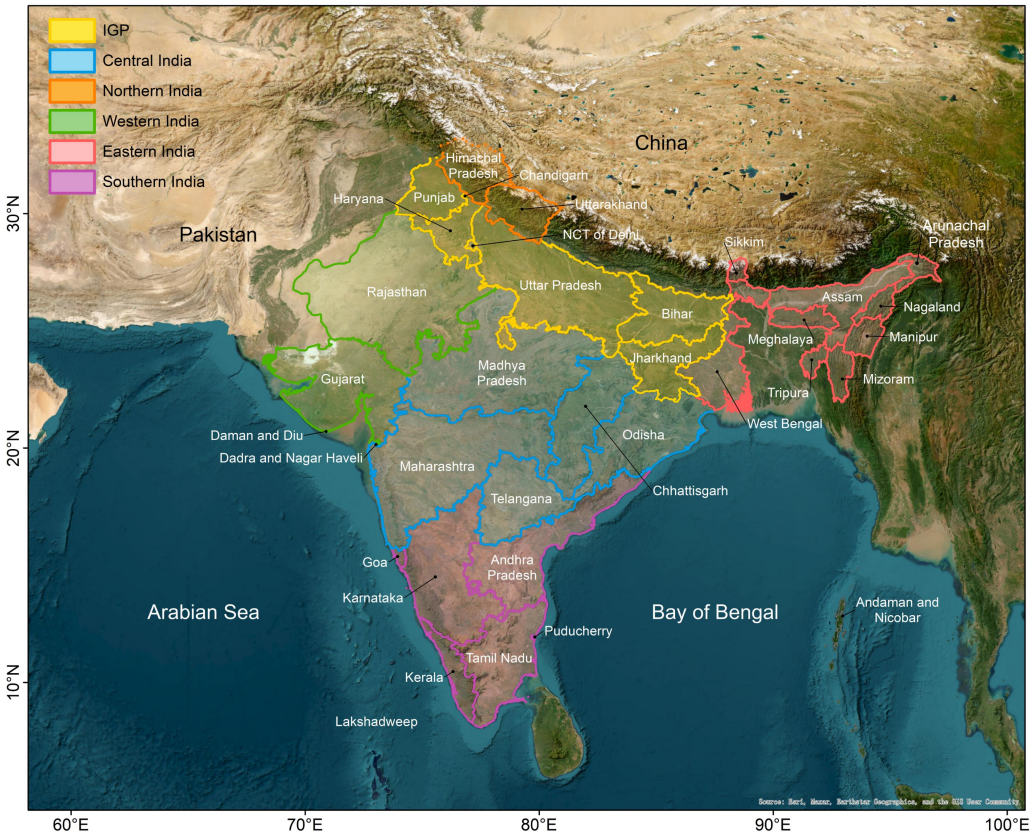

Figure 1. A map of India marked into six regions based on meteorological conditions and aerosol variability (adapted from David et al. 2018).

Please incorporate the methodology explained in Response 7 in the revised manuscript if not already.

Response: Thanks for your feedback. We have incorporated the methodology explained in Response 7 into the revised manuscript as follows:

Line 239: Hence, here we quantify the seasonal trend of $PM_{2.5}$ and O_3 from ANTHRO and BB emissions for DJF (December-January-February), MAM (March-April-May), JJA (June-July-August, monsoon season), and SON (September-October-November, post-monsoon season) from 1995 to 2014 by subtracting the BASE scenario from the FixAN or FixBB scenarios. The annual trends for $PM_{2.5}$ and O_3 for each season were subsequently estimated using the

Theil-Sen estimator and the Mann-Kendall test.

Line 266: These contributions are quantified by subtracting the BASE scenario in 2014 from the FixBB scenario in 2014.

Responses to Referee 2:

Response 1. The Line 14 is not grammatically correct and did not highlight the non-urban areas properly. Please revise. Also, what did they find in those earlier studies mentioned in Line 67? Does this study validate their findings? Please include in your result discussion.

Response: Thanks for your feedback. We have made the following revision:

Line 14: Anthropogenic (ANTHRO) emissions and biomass burning (BB) are major contributors to ambient air pollution, with the latter playing a particularly dominant role in non-urban regions.

Gurjar et al. (2016) demonstrated that anthropogenic sources are the primary contributors to SPM and PM_{10} in three megacities in India based on observational data. And, Vohra et al. (2022) found that increases in NO_2 in Indian urban areas were driven almost exclusively by anthropogenic sources, not traditional biomass burning, using both observational and satellite data. Our study, which covers both urban and non-urban areas in India, supports these findings as shown by the overall trend contributions. It confirms that changes in ANTHRO emissions were the dominant factor behind the deterioration of $PM_{2.5}$ and O_3 in the country, as stated in Line 224. To make it clearer, we revise line 224:

Line 222: Not surprisingly, changes in ANTHRO emissions dominated the deterioration of $PM_{2.5}$ and O_3 in India, consistent with previous studies based on both observational and satellite data (Gurjar et al., 2016; Vohra et al., 2022).

Response to methods. Please incorporate it in the revised manuscript if not already.

Response: Thanks for your feedback. We have made the following revisions to the manuscript:

Line 125: Both the Mann-Kendall test and Theil-Sen estimator require independence and randomness in the data, making them suitable for identifying monotonic trends.

Response to Interpretation Comment 2. Please incorporate it in the revised manuscript to improve the flow of your manuscript. Rather than start your discussions with "Figure x shows…", explain what/why you are going to do first. While you "concluded" something, the figures "show", not "showed", something, etc.

Response: Thanks for your feedback. We have incorporated it into the revised manuscript and clarify the context. as follows:

Line 221: To disentangle the contributions of ANTHRO and BB emissions to long-term trends in $PM_{2.5}$ and O_3 concentrations in India from 1995 to 2014, we first analyze their contributions to annual and seasonal trends (Figure 4).

Line 264: BB emissions exhibit a high degree of interannual variability, leading to less clear trends in the annual data. Thus, Figure 6 focuses on the spatial distributions of BB contributions for seasonal $PM_{2.5}$ and O_3 changes between 1995 and 2014 rather than trends, as detailed in Table S4.

Line 272: Therefore, despite their variability, the BB emissions in India posed a great threat to the air quality and thus could not be overlooked.

What do you mean by "to higher altitudes where the ozone lifetime is prolonged, facilitating accumulation"?

Response: Thanks for your feedback. Due to strong convection, ozone and its precursors are transported to higher altitudes, where ozone tends to remain in the atmosphere for a longer period because the rate of destruction is slower. As a result, ozone can accumulate at these altitudes. Here's the revised version:

Line 53: Additionally, strong convection enhances the transport of ozone and its precursors, such as NOy, to higher altitudes, where the prolonged ozone lifetime promotes accumulation.

For the grid choice, I understand the different lat lon for global study. However, this study conducted new simulations for the purpose of a regional study of India. Why do the authors still keep the same resolution and model domain as global modeling, especially given the availability of 0.5 degree resolution emission and other higher resolution data?

Response: Thanks for your feedback. This study is based on the results of global model simulations to provide a detailed analysis of India, which is why the grid resolution remains consistent with the global model. Future studies focused on India can use regional models with higher resolution to capture finer spatial details.

Please revise Figure S6 to grid cell maps, not administrative regions, to provide more spatial details.

Response: Thanks for your feedback. We have revised Figure S6 as follows:

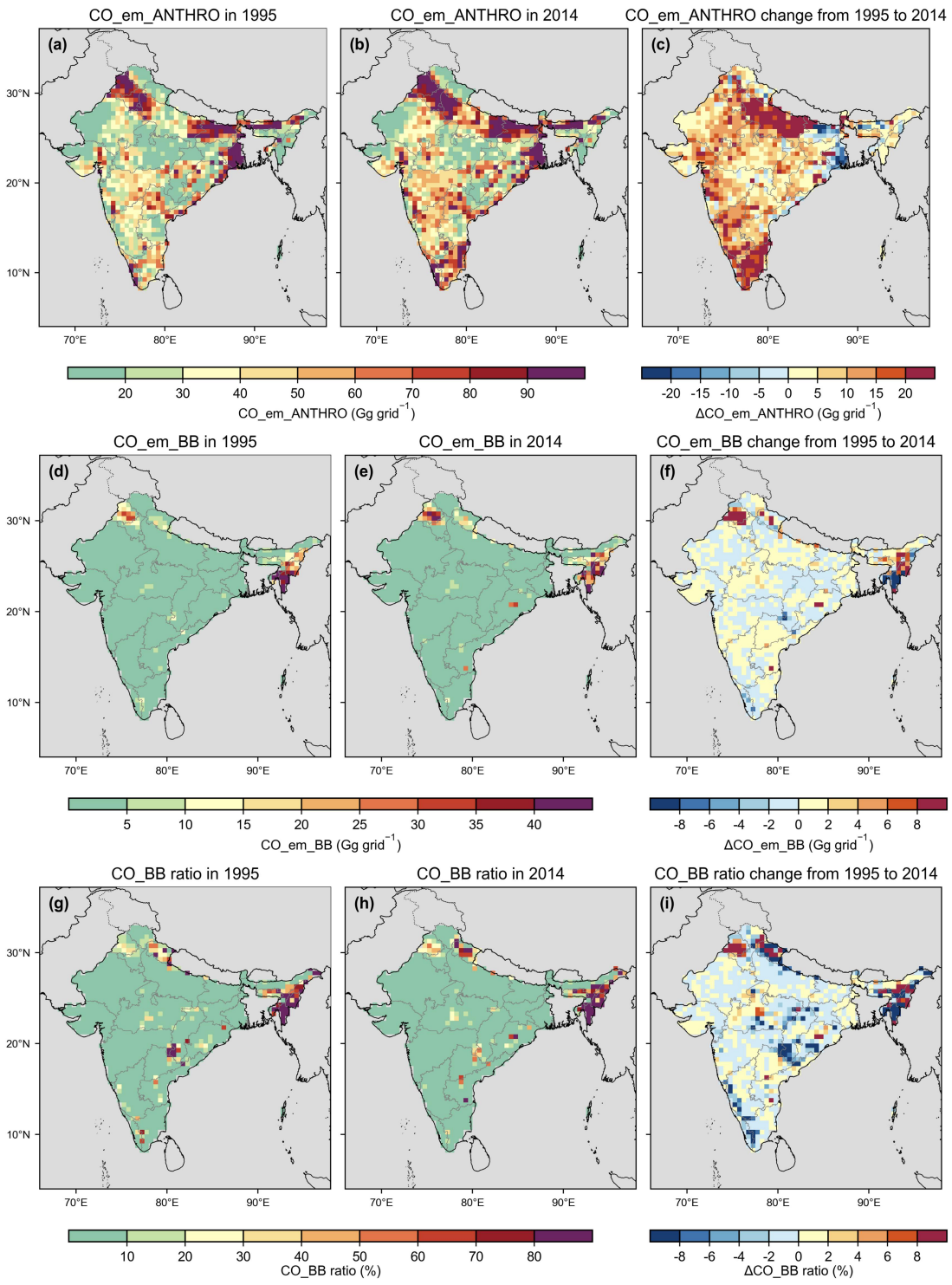

Figure S6. The spatial distribution of CO from (a) ANTHRO emissions, (b) BB emissions, and (c) the ratio of CO from BB emissions to CO from ANTHRO emissions in 1995 and 2014. The changes from 1995 to 2014 are also presented.

Let me rephrase my Comment 18: What uncertainties does the 95% CI in Figure 6 account for?

Response: Thanks for your feedback. The 95% CI is derived from the RR estimates of long-term exposure to $PM_{2.5}$ and O_3, as described in Section 2.4, which assess the correlation between long-term exposure to $PM_{2.5}$ and O_3 and the mortality burden from specific diseases. Two-sided

P-values based on the likelihood ratio statistic were calculated to assess the significance of these estimates (Turner et al., 2016). We have revised manuscript as follows:

Line 295: The shaded area indicates the range of 95% confidence interval account for RR estimates of long-term exposure to $PM_{2.5}$ and O_3 (gray indicates half of the range).

References:

Turner, M. C., Jerrett, M., Pope, C. A., Krewski, D., Gapstur, S. M., Diver, W. R., Beckerman, B. S., Marshall, J. D., Su, J., Crouse, D. L., and Burnett, R. T.: Long-Term Ozone Exposure and Mortality in a Large Prospective Study, Am J Respir Crit Care Med, 193, 1134–1142, https://doi.org/10.1164/rccm.201508-1633OC, 2016.

"Instead of India which could potentially have a higher value." is not clear and grammatically wrong. Please revise.

Response: Thanks for your feedback. After reviewing the relevant literature, we identified an error with the content. Here's the revised version:

Line 356: Meanwhile, in estimating the mortality burden, we apply the RR derived from a global study, rather than using values specific to India, which could potentially be lower (Brown et al., 2022). Thus, our estimations for the air pollution-related mortality burden could be overestimated.

References:

Brown, P. E., Izawa, Y., Balakrishnan, K., Fu, S. H., Chakma, J., Menon, G., Dikshit, R., Dhaliwal, R. S., Rodriguez, P. S., Huang, G., Begum, R., Hu, H., D'Souza, G., Guleria, R., and Jha, P.: Mortality Associated with Ambient $PM_{2.5}$ Exposure in India: Results from the Million Death Study, Environmental Health Perspectives, 130, 097004, https://doi.org/10.1289/EHP9538, 2022.

What do you mean by the following? "Finally, another limitation in our experimental design was that we set global fixed emissions for ANTHRO and BB instead of in India only, ignoring the impact of intercontinental transportation."

Response: Thanks for your feedback. The sentence refers to a limitation in the experimental design of the study. The FixAN or FixBB scenarios fix the global ANTHRO and BB emissions to 1995 levels, rather than focusing solely on India. Therefore, this study overlooks the impact of intercontinental transportation, such as how pollutant changes from other countries or regions might affect air quality in India.

Again, please check and correct all grammatical errors. E.g. "The ANTHRO emissions includes eight sectors" ...

Response: Thanks for your feedback. We have carefully reviewed manuscript to correct the grammatical errors. Now we revise it as following:

Line 91: The ANTHRO emissions include eight sectors: agriculture; energy; industrial; transportation; residential, commercial, other; solvents production and application; waste and international shipping (Hoesly et al., 2018).