Reviewer Report of Mogno et al., 2021

Mogno et al. present a comprehensive modelling study of seasonal distribution of particulate matter over the Indo Gangetic Plain (IGP), one of the world's most populated and polluted region. The manuscript is well-written and contains valuable results. This is a good overview paper on the drivers of air pollution in the IGP. However, the authors are requested to frankly acknowledge and openly discuss the large discrepancies between their simulation and observations, and additionally provide further description of their method of sensitivity analysis and its interpretaion.

1. Could you double check if the population of IGP is 400 million as per most recent estimates and perhaps provide a suitable reference?

2. L37: get rid of the extra "(June to September)"

- 3. Add a sentence on the limitations of using the 1D VBS scheme
- 4. L150: "Fasibalad" should be "Faisalabad"

5. Please confirm if emissions of all species have been added together to produce the plots in Fig 2.

- 6. L206: "components"
- 7. L270: "out" should be "our"

8. Emissions from 2010 have been used to simulate pollution in 2017-18. Comment on the nature of change in emissions that has occurred during this period (for example, you can consult McDuffie et al., 2020) and the expected changes in model results.

9. Provide your reasoning/explanation why your model results have reported higher PM2.5 levels over lower IGP

10. L281: You mention that "post-monsoon biomass burning do not impact the central and lower IGP". However, as per Figure 4h, if I read the colorscale properly, there's considerable influence of biomass burning (pyrogenic) emissions – on the scale of 250-500 ugm⁻³ Gg⁻¹ all the way up to eastern Uttar Pradesh. These values appear to be higher than those from anthropogenic emissions in Figure 4g. Also, how do you reconcile this result with, for example, Ojha et al., 2020, who reported up to 20% contribution of biomass burning emissions to PM_{2.5} over central IGP cities? Since you are comparing values across different emission types, I suggest replotting Figure 4 and 7 using a common colorscale for all three emission categories.

11. Section 2.2: This section can benefit from a clearer description of the sensitivity analysis. For example, it is not clear what is Δt here – is it equal to a week, i.e., 7 days x 24 hours = 168 terms that were summed for each species for each gridpoint? Also, how is this particular sensitivity index supposed to be interpreted when thinking about control policies? It seems to me that you'd get a higher value for a grid if it is highly influenced by its own (local) emissions while other highly polluted grids may be influenced by emissions from several other grids leading to a much higher pollutant concentration but they won't show higher sensitivity values in this particular metric that you've

presented. This can cause confusion in interpreting such a metric especially for policymaking, when pollution is highly regional as is the case in IGP. Therefore, I request you to provide a richer discussion on the meaning and interpretability of the sensitivity index maps presented in Figure 4 and 7.

12. L352: "Punjabi Pakistan" would be better replaced by "Pakistani Punjab"!

13. Figure 8: Please increase size of the legend (gas,aerosol)

14. L400: $log_{10}C^* = 2$ is written twice here. Should it be = 3 in the second instance?

15. L452: Replace "then" with "than".

16. Table A3: The model shows poor performance for NO₂ all year round. Given that nitrate aerosol is a significant portion of PM_{2.5} as simulated by your modelling system particularly during post-monsoon and winter, how confident are you about this result? How does it compare with field observations such as Gani et al, 2019; Patel et al. 2021; Gunthe et al., 2021? Similary, how do you explain the dramatic overprediction of SO₂ during post monsoon and winter and such low r-values? What are its implications for the simulated sulfate in your model? How do you explain the negative r-values for Ozone all year round? What are its implications for SOA simulation and regional contribution? These model weaknesses need to be acknowledged and discussed and the results and conclusions of the study need to be qualified in light of these weaknesses.

17. L351: You say that OA distribution during post-monsoon is most sensitive to changes in anthropogenic emissions but if I read the colorscales correctly, the sensitivity index is a maximum of 100 for anthropogenic emissions but is way above 100 for biomass burning emission over large parts of IGP. Again, please replot this figure with a common colorscale and reinterpret.

18. Figure 4 and 7: During the monsoon season, the sensitivity of $PM_{2.5}$ and OA is higher towards changes in biogenic emissions than to changes in anthropogenic emissions. The reasons for this should be detailed in the text.

19. L299: Here you attribute the high PM_{2.5} concentrations in the lower IGP during pre-monsoon primarily to meteorology but this is actually a peak biomass burning season over the Myanmar-Laos region (see, for example, Figure 3 and 5 in Ansari et al., 2016; Figure 5 in Reddington et al., 2019). You can check your pyrogenic emissions file if it contains emissions beyond lower IGP, otherwise it must be coming from the chemical boundary conditions. You should include this influence in the sentence.

References:

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Gunthe, S.S., Liu, P., Panda, U. et al. Enhanced aerosol particle growth sustained by high continental chlorine emission in India. Nat. Geosci. 14, 77–84 (2021). <u>https://doi.org/10.1038/s41561-020-00677-x</u>

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Reddington, C. L., Conibear, L., Knote, C., Silver, B. J., Li, Y. J., Chan, C. K., ... Spracklen, D. V. (2019). Exploring the impacts of anthropogenic emission sectors on PM\$_{2.5}\$ and human health in South and East Asia. Atmospheric Chemistry and Physics, 19(18), 11887–11910. https://doi.org/10.5194/acp-19-11887-2019

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