

Review of Source influence on emission pathways and ambient PM_{2.5} pollution over India (2015-2050) by Venkataraman et al.

This study developed scenarios of sectoral emissions of PM_{2.5} and its precursors for 2015-2050 and further assessed the impacts of individual source-sectors on PM_{2.5} pollution through GESO-Chem model simulations over India. Based on model simulations authors have shown that under the present day emissions most states in India exceed NAAQ standard of 40 µg/m³ (annual mean). Based on emission evaluation under proposed regulations authors have shown further deterioration of air-quality in 2030 and 2050, even in highly ambitious scenario 10 states in India will not meet the current NAAQ standard in 2050. Overall, their finding suggests that residential biomass burning and agricultural residue burning is the primary largest sector (highly uncertain sector and not validated with the in-situ data) contributing to the large regional background of PM_{2.5} pollution in India. The paper presents interesting analyses and will be an important resource for the community. However, I have some queries given below and certain key issues need to be addressed for improving the discussion section before it can be accepted for publication. Please find some suggestions below which I hope the authors may find useful for revising the MS for improving the discussion on the issues that affect the uncertainty/certainty of present findings and conclusions.

My major concern is lack of sufficient validation/evaluation of the capability of a well-respected model to simulate chemical species over India, a region with limited publicly available observations. These are very important for meaningful future research too as PM_{2.5} is a pollutant derived from several precursor emissions with varied sources. Currently the work does not acknowledge such issues and puts too much stock by the model results. Even the model was previously applied to study PM_{2.5} over India relating satellite AOD to ground-level PM_{2.5}, there has not been a great deal of comparison of model results against observations in previous studies. Global off-line models have large difficulties in simulating chemical species over India (Surenderan et al., 2015, 2016 AE). Therefore it is essential to build confidence in the ability of GEOS-Chem model (since it is finest resolution) to simulate species distributions reasonably well so that it can be used for sensitivity simulations (such as performed for this study) and to understand future air quality projections. Large biases in model may influence the regional PM_{2.5} fields in the future projections which I believe make it difficult to draw conclusions that are of scientific value. The authors should clearly address this point by comparing the model with the observed PM_{2.5} for greater understanding of model biases and recognition of areas needing improvement.

As a part of evaluation work for HTAP-II PM_{2.5} and BC data (mostly from the published literature (not necessary for the same year)) has been compiled for more than 15 stations in India which can be shared to the author for model validation. Of course, I cannot categorically state that there is a problem, but I do find in figure 4 & 5 that the model has difficulties in simulating the species distribution.

There is always a problem of representativeness when comparing coarse-scale models to point observations and perhaps this could be a problem. I would also suggest to the authors to

review how they have compared the simulated PM_{2.5} (model lowest level??) with in-situ observations and satellite AOD (model field interpolated to satellite overpass time).

Are NH₃ emissions fixed to 2015 level in BAU, S2 and S3 scenarios? NH₃ is important compound for the formation of secondary aerosols and agricultural activity is one of the major sources of NH₃ in India, particularly in the rural India where residential bio-fuel and biomass burning is dominant. It is necessary to clarify how authors have treated NH₃ in 2015 and further in BAU, S2 and S3 scenarios. Considering projected growth in agricultural sector in India it is believed that NH₃ emissions will increase further (Sutton et al., 2017). Therefore, it may have some implication on future PM_{2.5} levels.

Second concern:

It is understandable that due to lack of primary measurements concerning several important emission types (e.g. NMVOCs), the magnitude of these emissions are still poorly constrained in the emission inventories and are yet to be validated using in-situ data or with representative emission factors determined from measurements conducted within India from major sources. However, it is necessary to highlight these existing uncertainties arising from the data limiting factors and which are currently substituted through use of emission factors that may not be representative of emission sources in the South Asian atmospheric environment.

1) The authors should provide a speciated list (even in supplement would do) for the NMVOCs considered in this work. Individual NMVOCs have different PM formation potential and without such information it is not possible for the reader to assess how well this class or precursor has been constrained.

2) The key finding reported by the authors concerns the major contribution due to the emissions from traditional biomass technologies in the residential sector (for cooking and heating), the informal industry sector (for brick production and for food and agricultural produce processes), as well as from agricultural residue burning. (Lines 17-20; Page 4 of MS). In this regard, it is necessary to point out several recent studies conducted in Nepal (see Special issue in ACP on Atmospheric pollution in the Himalayan foothills: The SusKat-ABC international air pollution measurement campaign Editor(s): S. S. Gunthe, E. Weingartner, K. O. Nguyen Thi, and E. Stone) and in particular the following papers: Stockwell et al., 2016 and Sarkar et al., 2017). Stockwell et al conducted rare, field measurements in South Asia of emission factors for up to 80 gases (pollutants, greenhouse gases, and precursors) and black carbon for many previously under-sampled sources that are important in developing countries such as cooking with dung and wood, garbage and crop residue burning, brick kilns, motorcycles, generators and pumps, etc. The authors should discuss this work in some detail and compare the emission factor values for reported sources with values used in their work and shown in Table S7. This is important to gauge how much uncertainty can arise from use of variable emission factors. Secondly the work by Sarkar et al. 2017 provides valuable insights on where current emission inventories need to be improved for better representation

of emission source contributions. It provides quantitative information regarding the source contributions of the major NMVOC sources in the Kathmandu Valley. Combining high-resolution in situ NMVOC data and model analyses, it showed that REAS v2.1 overestimates the contribution of residential biofuel use and industries. This is very pertinent to discuss and include in the context of the present work for the following reasons. The use of emission factors from residential biofuel sources for determining ambient source contributions without adequately accounting for the deposition and/ or other loss that can occur for the indoor emissions due to household cooking/heating and their net emission to outdoor environment can lead to gross over estimation of the emissions as an atmospheric source. The results of Sarkar et al., 2017, which is focused on NMVOCs appear to point towards such loss processes being significant and if true, this is likely to be even more important for PM_{2.5} that has higher deposition tendency than gases. These important aspects need to be highlighted and addressed so that future work can benefit from such insights. Are there any similar NMVOC datasets reported from the Indian region? It would be good for the authors to mention these if possible. For many of the biomass burning sources, it is now recognized that combustion efficiency can be even more important than the fuel composition for the emission factors (Roden et al., 2006; Martinsson et al., 2015). Recent relevant work on open agricultural stubble fire emissions of NMVOC from north-west India (Kumar et al., 2018) which appeared after the present work was already in ACPD, may also be helpful for discussing issues pertaining to the inadequate accounting of all gaseous organic gases and uncertainties concerning emission factors.

Minor issues:

Page 10, line 30: 'open burning were derive from the global GEFD-4s database' This statement suggests that the authors have used both GEFD-4s open burning emissions as well their own estimated biomass burning emissions for 2015, BAU, S2 and S3. How different GEFD-4s open burning is from the open burning assessed in the present work? Authors should clearly address this point.

Page 13, lines 25-30: I have some reservations about the statement made here because sectorial emission distribution is so diverse in India that some regions may see significant change in air quality even in S2 scenario but not necessarily as a regional mean. I would welcome a figure with summary statistics about PM_{2.5} concentrations for BAU, S2 and S3 scenario for 2105, 2030 and 2050 (e.g., box-whisker plots mean, median, standard deviation, and P25, P75).

Page 14, line 15: The term population weighted mean PM_{2.5} concentration needs to be defined.

Page 14, line 28: open burning (agricultural) again how different it is from the GEFD-4s? Pl. make sure that it is now counted double.

Page 17, line 7: Is expansion in industrial process in assumed at the same grid locations in BUA, S2 and S3 scenario? If yes, please mention it categorically.

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