

Review of “Why the Indo-Gangetic Plain is the region with the largest NH₃ column in the globe during summertime?”

General Comments

This study aims to explore the reasons behind the elevated levels of ammonia observed over the Indo-Gangetic Plain. This is an important and scientifically relevant question, particularly since the ammonia burden has significant implications on inorganic aerosol concentrations over the region. The authors use the WRF-Chem model to investigate the physics and thermodynamics underlying the atmospheric fate of NH₃ and the resulting analysis provides useful insights into some of the factors driving the high concentrations over the region. For instance, the demonstrated sensitivity to increasing NO_x and SO₂ emissions is an interesting result, particularly when contrasted to the case over the North China Plain.

While the dynamic physical transport and meteorology simulated by the model is validated and constrained, the approach chosen for the thermodynamic analysis largely hinges on an offline test of emissions and temperature sensitivities using the ISORROPIA model. The analysis would benefit significantly from a more rigorous on-line treatment of the thermodynamics that investigates the specific factors controlling NH₃ partitioning in greater detail. In addition, observational constraints (such as satellite ammonia columns) could be further leveraged to get a more quantitative estimate of model performance prior to interpreting the model output. With this in mind, I provide the following comments below and recommend that these issues be addressed prior to publication in ACP.

Specific Comments

Model Details, Validation and Uncertainties

In order to appropriately interpret the WRF-Chem analysis the authors should provide a more detailed discussion on the specifics of the ammonia simulation in WRF-Chem. For instance, is it possible to run ISORROPIA in an online configuration (partitioning at each time-step and explicitly simulating the aerosol species)? Unless I am misinterpreting the methods section, it appears that ISORROPIA is used only in an offline context. Given the spatial and temporal heterogeneity in the various factors that drive aerosol partitioning, running ISORROPIA in an online configuration would more appropriately explore the scientific questions outlined in this study.

Line 70: The authors have validated the simulated meteorology (wind speed, temperature, etc.) but do not validate the ammonia simulation itself. While I recognize this is challenging, it could potentially be done using satellite measurements (with the appropriate application of an averaging kernel) or surface measurements where available. Even a general estimate of how well the model captures ammonia variability and magnitude over the region would provide important context. Ideally, the different (non-transport) factors that dictate ammonia concentrations (namely – emissions, wet deposition, dry deposition and aerosol partitioning) would be constrained using observational data whenever available. In absence of such data, an explanation of the uncertainties associated with these various processes (and the steps that need to be taken to constrain them) is required in order to appropriately interpret the results.

Line 53: The simulations described here are spun-up over a 7-day period. A more detailed discussion about the initial concentrations assumed for the most important chemical species and their estimated

lifetimes would provide useful context on whether the week-long period provides sufficient time to allow the longer-lived gas-phase species to equilibrate prior to the main simulation period.

Seasonal Analysis

Line 52: The authors classify June to August as the summer period. However, in India, this season is characterized by the monsoons (usually beginning in mid-June) which are associated with drastic changes to regional meteorology. This perhaps provides context for the statement on Line 198, given that high levels of precipitation and humidity are expected during the monsoon season. The Indian summer is usually thought to be between the months of April – May.

Impact of Transport and Meteorology

Section 3.2 discusses the importance of various meteorological drivers (such as RH and temperature). However, given its importance in determining ammonia burdens, a more detailed discussion of the specific mechanisms that dictate aerosol partitioning under different meteorological conditions and the associated uncertainties in our understanding of these processes would add to the broader utility of this study.

Use of the ISORROPIA module to assess the impact of emissions and temperature

Line 77: A more detailed overview of the ISORROPIA module would provide important context for the resulting analysis. If only applied in an offline context, it is possible that the analysis is not capturing various important (and non-linear) effects due to the spatial heterogeneity in gas phase and particle concentrations (along with the associated depositional losses at every timestep). While the authors provide an observational constraint (Line 142) to validate this approach, the differences in the model and observed partitioning ratio are significant (on the order of 30%). A more thorough comparison with the observational data would greatly benefit the analysis and serve as validation for some of the later conclusions. Additionally, the comparison of the regional mean to observational data over Delhi may not be appropriate, particularly given that NO_x and SO₂ concentrations are likely much higher over the city.

Line 155: The temperature sensitivity is an interesting result, particularly when contrasted to the SO₂/NO_x sensitivity. However, the approach here considers only a simplified case over the entire region. If the partitioning was conducted online (at every timestep), would it be reasonable to expect a different sensitivity to changes in temperature / SO₂ / NO_x? A more detailed discussion about the non-linear, spatially dependent factors driving aerosol concentrations (and the heterogeneity in emissions, loss processes, thermodynamics, etc.) would provide more context with which to interpret these results.

A discussion of other drivers of aerosol formation (particularly in the context of the monsoons)

Singh and Kulshrestha (2012), cited in this study, hypothesized that humidity during the monsoon season had a significant impact on NH₃ partitioning. Could the authors discuss this in the context of their analysis? Given that the aim of this study is to establish the most salient drivers of the high NH₃ concentrations (particularly during the selected monsoon period), a more detailed discussion of what determines the relative dominance of the different production, loss and partitioning mechanisms under various atmospheric conditions would provide important context with which to interpret the results of this analysis.