

Review Report:

Title: *Optimizing Ammonia Emissions for PM_{2.5} Mitigation: Environmental and Health Co-Benefits in Eastern China*

Authors: Keqin Tang et al.

This manuscript presents an inverse modeling framework that combines IASI satellite observations with the WRF-Chem model to optimize ammonia (NH₃) emissions over Eastern China. The results suggest that prior inventories substantially underestimate NH₃ emissions—by approximately one-third for the year 2016. The study captures strong seasonal variability, identifies the dominant role of agricultural sources in summer, and highlights the importance of non-agricultural sources in winter. It also evaluates the downstream benefits of NH₃ mitigation on PM_{2.5} reduction and public health.

This is a timely and policy-relevant study with important implications for emission inventories, air quality management, and health co-benefits. The manuscript is generally well organized and written, but certain aspects of the methodology, validation, and interpretation require clarification to ensure scientific rigor and transparency.

I recommend **major revisions** before the manuscript is accepted for publication.

Major Comments

1. Limited Optimization Period:

- The top-down optimization was performed only for four months (each season). Please justify why the analysis was limited to these periods and discuss whether this may affect annual emission estimates or bias seasonal interpretations.

2. Validation with Surface Data:

- While Figure S2 attempts to demonstrate agreement between model and surface observations, a time series or seasonal comparison between observations at these sites and both prior and posterior simulations would provide more clarity.
- Consider including monthly or seasonal cycle plots at selected sites to demonstrate how well the posterior simulation captures temporal variability.
- Scatter plots comparing prior vs. observed and posterior vs. observed NH₃ concentrations (similar to Figures S4–S6) should be more clearly explained in the main text.

3. Clarification on Posterior Emission Totals:

- Page 13, L343: Clarify whether the 4.2 Tg emission is derived from the posterior estimate. Given the remaining model–observation gap, this number should be framed as a lower-bound estimate. Please discuss the implications.

4. Sectoral Emission Trends and Inconsistencies:

- Page 8, L216–218 and L223: There seems to be a discrepancy between the statements about non-AGR and AGR emission changes. Figure 5b suggests a spring increase, typically associated with agricultural activity, yet the larger change is attributed to non-AGR sources. Please clarify the partitioning of the emission increase.

5. **Loss of High Emission Feature in SON/DJF (Figure 6):**
 - The high-emission feature at the intersection of Henan, Hebei, and Shandong disappears in SON and DJF seasons. Please explain whether this is due to real seasonal changes or limitations in the model/data.
6. **Surface Bias Reduction (Page 10, L277–278):**
 - Clarify which observational data were used (in situ surface measurements vs. satellite), whether the bias reduction is spatially averaged, and if supporting plots are available. A comparison showing seasonal variation would strengthen this claim.
7. **Table 3 Description (Page 11, L288–297):**
 - Provide more detail on the meaning of the single values listed in Table 3. What metrics are these? How do they compare across seasons and sectors?
8. **Public Health and PM_{2.5} Impact:**
 - Page 11, L306: Quantify the reduction in PM_{2.5} (1.5–5.7 µg/m³) as a percentage of baseline concentrations to help contextualize the health impact.

Minor Comments

- **Page 4, L97:** Surface data usage should be mentioned in the abstract for completeness.
- **Page 7, L164:** Briefly explain the extreme values of IOA and MFB to aid reader interpretation.
- **Page 7, L180–184:** Confirm whether these lines are referencing Figure 6.
- **Page 8, L201:** Remove extra period after “Table 2.”
- **Page 9, L239:** Correct grammar: "based on both top-down and bottom-up approaches."
- **Page 10, L265:** Suggest adding the prior result value alongside the percentage difference for clarity.
- **Page 11, L281:** Reword the statement to reflect partial improvement in posterior vs. surface observations.
- **Page 13, L342:** Add missing period.
- **Page 13, L348:** Consider providing a geophysical or socioeconomic explanation (e.g., dense agriculture and livestock) for high emissions at the provincial intersection.

Suggestions for Figures

- **Figure 2:** Add a row showing differences (posterior – prior) for better visualization of emission changes by sector.
- **Figure 3:** Clarify what the red box highlights; also discuss large underestimates in Shandong and BTH.
- **Figure 6:** Add units to the color bar for clarity.
- **Figure S2:** Increase font size on color bar units.

References

- Please ensure that all relevant IASI-based ammonia studies are cited appropriately to position your work in the broader context of satellite-based NH₃ retrievals and applications.

Conclusion

This is a strong contribution to understanding ammonia emissions and their health and environmental implications in a rapidly developing region. The study is scientifically valuable, but several key clarifications and visual enhancements are needed to strengthen its conclusions and reproducibility. I recommend revision with attention to the above points.