Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2021-549-RC1, 2021

Comment on acp-2021-549

Anonymous Referee #2

Referee comment on "An ensemble-variational inversion system for the estimation of ammonia emissions using CrIS satellite ammonia retrievals" by Michael Sitwell et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-549-RC1, 2021

- **Summary**

Estimating ammonia emissions is often challenging due to timing, method, and amount of fertilization varying spatially and temporally over different regions and crop lands. In recent years, ammonia retrievals from different satellite platforms became available. Improving ammonia emissions using satellite retrievals which have better spatial and temporal coverage seems to be promising. The goal of this paper is to estimate ammonia emissions using an ensemble-variational inversion approach with Canadian GEM-MACH chemical weather model and ammonia satellite retrieval from the CrIS instrument aboard on the S-NPP satellite. The inversion is conducted from May to August 2016 and the results are evaluated against surface observations. The research approach is well described in the paper and the content is well organized overall. Results are presented in many ways through model evaluations against surface observations for many different species. However, the results presented throughout the paper are mainly limited to the normalized mean bias (NMB) metric, which could be misleading on average without also considering absolute error metric evaluation.

- **Comments**

Two areas are identified for major improvements in order to be accepted for publication.

- **Evaluation**. Throughout the results, total NMB (e.g. Figures 7, 11, 13, 14,16) from all sites are used to demonstrate the impacts of the updated ammonia emissions using the inversion approach. Mean bias evaluation can be misleading without also evaluating absolute error (ME or RMSE) due to the possibility of positive and negative biases

canceling each other RMSE is presented in the supplement figures (Figures S3, S5, S6, S7, S8) and it seems that many sites show worse performance. However, model performance related to RMSE evaluation is not mentioned in the main paper at all. It is important to evaluate the updated model using bias and error metrics together to understand the influence of the inversion approach. RMSE should be presented in the paper with NMB figures (7, 11, 13, 14, 16).

- **Sensitivity of constants**: More discussion should be given on model performance spatially in relation to the inversion formulation. As demonstrated in the paper, observation operator selection (log, linear, or hybrid) greatly influences the model performance. For instance, based on Figure S3, sites in the western central U.S. (around Colorado) tend to have worse performance. Could it be related to the constants used for cut-off in the linearized observation operator given that this area tends to have low surface emissions (based on Figure 9)? Sensitivity analysis on the constants used in the hybrid approach seems to be important and useful for the "ideal" constant selection. Maybe the values used for linear-log cutoff should be variable spatially or temporally depending on the ground sources. For the novel hybrid approach developed, one important question to address is how the model is sensitive to the selected constant values in the hybrid approach over such large domain. Instead, it seems that much discussion and explanation are given to rationalizing the high biases in the fine and coarse PM estimations.

More specific minor comments are listed below:

- 92 line: what does "The number of degrees of freedom for this retrieval is 0.956" mean?
- May to August 2016 study. Since this approach is developed for the GEM-MACH air quality forecasting model, probably it is important to evaluate how this approach performs in other seasons with cooler temperature and low ammonia emissions as well.
- In reality, fires exist, and fire emissions are included in forecasting. Is this approach appropriate if weekly updates are applied for emissions under fire conditions?
- 98-103 lines: What is magnitude of the issue related to the non-detection of ammonia discovered on the quality of CrIS data which affects non-source regions in the domain?
- 109 line: 70–85% of the retrievals used in the inversions coming from daytime retrievals. What causes nighttime retrievals to have low quality?
- Figure S1: hard to tell difference among 0 to 50 color scale in plots.
- Figure 9a – NH_3 value higher than graph horizontal range.
- Why do all RMSE (updated) / RMSE (original) ratio figures (S3, S5…, S8) in supplement have negative values?
- The inclusion of the critical load exceedances seems to be out of the focus for this paper although updating ammonia emissions affects N deposition. Given the purpose of the paper and more thorough evaluation needed for this new approach, it is recommended to remove the critical load results from the paper.