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Atmospheric Chemistry and Physics Discussions

Interactive comment on "High-resolution Hybrid Inversion of IASI Ammonia Columns to Constrain U.S. Ammonia Emissions Using the CMAQ Adjoint Model" *by* Yilin Chen et al.

Anonymous Referee #1

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This manuscript by Chen et al. used the recently developed multiphase CMAQ adjoint model and IASI satellite total NH_3 column observations to constrain the monthly NEI NH_3 emissions at 36 km spatial resolution in April, July, and October in 2011. A hybrid, two-step optimization scheme was applied. First the NEI inventory was nudged towards the posterior values by a mass-balance approach at a much coarser grid (216 km), and then 4D-Var inversion was performed using the updated inventory as the prior. The posterior emissions were then used to drive the CMAQ model, and the simulated NH_3 abundance, NH_4 deposition, and aerosol chemical composition were evaluated against independent observational datasets. Overall this work is solid, has applied state-of-the-art satellite data and CTM tools, and could advance our limited understanding on the

emission of NH_3 if its methodology can be fully justified. Hopefully the paper can be further improved after addressing my comments below.

General comments

1. NEI 2011 covers the entire year continuously but this work only focused on three months, April, July, and October. Presumably the computing cost prohibited optimizing NEI for other months, but this should be discussed. Many CTM users would use multiple months or the entire year of NEI, and those three isolated months would hinder further application of the results of this work. Especially, the month of May will be a significant opportunity missed as a large fraction of fertilizer application happens in May, leading to abruptly different emission and column density dynamics relative to April and June.

2. The observation used in the inversion seems to be monthly averaged data over 36-km grid cells, and the grid average absolute error was used in the observational error covariance matrix. This may have led to the counterintuitively high values in Pennsylvania and southern Texas, as the monthly averaged grid value could have been driven by a few anomalously high observation dates, given the sparsity of IASI pixels. The error term (in Equation 1) does not include the scaling of the square root of N (the central limit theorem). As a result, if a grid cell contained only one day with extremely high values (the other days in the month were missing), it would be treated the same way as if all 30 days were those high values. Specifically, the high emissions in Pennsylvania, western New York, and east/south Texas (Fig. 3d) that were seemly driven by high IASI values in April (Fig. 1a) are hard for me to believe. It might be helpful to check IASI April data in other years, e.g., 2010 and 2012, to see if those high column abundance (and consequently high posterior emissions) are consistent.

Specific comments

Page 2, line 49: clarify which NEI it is (prior or posterior) in "NEI-based" assessments.

Page 2, lines 61-65: this sentence might fit better at the last paragraph of the introduction.

Page 5, equation 1: this is a strange statistic to calculate. As indicated a few lines above, Ω_0 is the monthly arithmetic mean within a grid cell, but the $\sum(\sigma_i/\sigma_i^2)/\sum(1/\sigma_i^2)$ term is the variance-weighted mean of error. A simple standard error of the mean or standard error of the weighted mean (https://doi.org/10.1016/1352-2310(94)00210-C) might be better choices.

Page 7, lines 201-202: how justified is it to assume that the a priori covariance matrix is diagonal? The error/bias in NEI often seem spatially correlated.

Page 7, lines 203: it is important to let the readers understand if the observation vector used in the inversion is composed of single IASI pixels (level 2) or regridded maps (level 3). My impression is that the level 3 regridded IASI data were used. In that case, the single sounding detection limit of 4.8×10^{15} is not relevant as the averaging will reduce the noise level, and it is important to consider the number of averaging per grid cell.

Page 7, line 215: the convergence criterion that J decreases by less than 2% seems large and arbitrary.

Figure 2: please consider adding the residual map (IASI column-modeled column) as inserts, similar to Fig. 1.

Page 8, lines 234-235: please define the exact location of NRMSE that reduced by 98%. The high NH_3 observations in April in southern states seem curious and may warrant a closer investigation.

Page 9, line 277: it may be helpful to also include a priori emission totals in those three months. The posterior emission indicates that the total NH_3 emission decreases linearly from April to July and to October. Then what would the seasonality look like?

Page 9, line 297 and page 12, line 384: it is contradictory to claim that the hybrid inver-

sion “overcomes the over-adjusting problem for high emission rates” and meanwhile attribute the worsening RMSE against AMoN to the emission over-adjustment problem that has supposedly been overcome. Especially the comparison between posterior and AMoN in April (Fig. 4a) seems problematic.

Table 1: the R^2 of 0.08 at other (also the majority of) sites between simulated NH_4^+ and observations in April is bothersome. The N is a reasonably large number (115), so such a low R^2 indicates that the model essentially lost all explanation power after the inversion. The authors are suggested to take a closer look at the April data (for other years than 2011 as well) and make sure they are representative.

Page 10, line 302-303 and page 11, lines 345-346: as CMAQ is a full 3D CTM driven by real WRF meteorology and hourly emissions, those transport should have been captured. Why not?

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