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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Inverse modeling and mapping US air quality influences of inorganic $PM_{2.5}$ precursor emissions using the adjoint of GEOS-Chem" *by* D. K. Henze et al.

D. K. Henze et al.

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We appreciate the comments of Reviewer #3, which we address both below and in our revised manuscript.

1. *I would have liked to see a robustness analysis of the converged solution; this can be done by repeating the inversion with different initial conditions.*

   While a comprehensive robustness analysis of every parameter in every season is computationally prohibitive, we have performed a robustness analysis for our inversion during July, repeating the inversion from several different initial estimates of the NH_3 emissions. The results indicate that the inversion places

an upper-bound on the NH_3 emissions during that month, while lower NH_3 emissions can not be ruled out. These additional test are now introduced at the end of Section 4.4:

"To explore the possibility that the local minimum found during the optimization is not the global minimum of $\mathcal{J}$, additional optimization tests can be performed starting with different initial guesses for the emissions scaling factors. To demonstrate, the optimization is repeated for July using a range of initial guesses for NH_3 emissions. The results from these tests are presented and analyzed in the following section."

The discussion in section 4.5.1 includes an additional figure (now Fig. 11) and the following text:

"Figure 11 shows the optimization results for anthropogenic NH_3 emissions during the month of July using a range of initial guesses for NH_3 emissions. Results for the standard optimization (initiated with $\boldsymbol{\sigma}_a$ = 0.0) are compared to results that begin with the following factors: $\boldsymbol{\sigma}_a$ = 0.69, 0.41 and -0.69, which correspond to doubling, increasing by 50% and halving the emissions, respectively. The results demonstrate consistency of certain features across each optimization test. Most visibly, the scaling factors in the south-central US are always -1.0 or less. Scaling factors stretching from Michigan to New York are between -0.5 and -0.3, even when obtained by increasing emissions in those area from the test that began with $\boldsymbol{\sigma}_a$ = -0.69. While some cells are estimated to have a positive scaling factor in some tests but negative scaling factors in other tests, the ranking of emissions adjustments in these cells relative to other areas within the same optimization test are mostly similar regardless of $\boldsymbol{\sigma}_a$. Exceptions are locations such as Southern California, where emissions increased when $\boldsymbol{\sigma}_a$ = 0.0 and $\boldsymbol{\sigma}_a$ = 0.69 but not when $\boldsymbol{\sigma}_a$ = 0.41. The final cost function is actually

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lowest for the σ_a = -0.69 test, for which $\mathcal{J} = 1366$. The cost function for σ_a = 0.0, 0.41 and 0.69 is 1469, 1615 and 1789, respectively. For the latter two tests, the total optimized US NH_3 emissions are greater than or equal the σ = 0.0 emissions. For the σ_a = -0.69 test, the optimized US NH_3 emissions were 53% lower than the σ = 0.0 emissions. Given the cost function values, it is most likely that the optimization results presented for σ_a = 0.0 in July are an upper bound on total US NH_3 emissions, which may be even lower, but not likely higher. This is reasonable given that observed nitrate aerosol concentrations are typically very low during the summer. Any exception to that would require much higher NH_3 emissions, but any number of lower NH_3 emissions are plausible as long as the nitrate concentrations remain within the range of model and instrument uncertainty."

2. *The representational error is assumed to be 30%, and is used to characterize the spatial (subgrid) heterogeneity in each grid cell. How was this value selected? Does it (and should it) also account for temporal variability as well?*

   The representational error was not implemented with temporal variability in mind. While important for comparison of model results to high frequency observations, such as plane flights or satellite measurements, temporal representational error is probably minimal for comparing the model to daily or weekly averaged measurements. Nevertheless, the reviewer brings up a good point; the choice of 30% representational error was somewhat arbitrary. This is now justified by additional studies to demonstrate that a choice of 10% or 50% does not greatly impact the inversion results; since the error is assumed to be diagonal, this only shifts the balance of the two terms in the cost function. Picking a different representational error would lead to a different, compensating,

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regularization parameter. These additional studies are now included in the revised manuscript in the second to last paragraph of section 4.4:

"The representational error that contributes to S_{obs}^{-1} also affects the optimization. ...When assumed to be a uniform fraction of the observed value throughout the model, the representational error only affects the balance between $J_{\text{prediction}}$ and $J_{\text{parameter}}$. For example, with 30% representational error and γ_r=10, the cost function in July reduces by 28%, and the nitrate RMS error reduces from 0.67 μg/m^3 to 0.44 μg/m^3. Using 10% (50%) representational error, the cost function reduces by 33% (26%), and the nitrate RMS error reduces to 0.43 μg/m^3 (0.45 μg/m^3). Repeating the regularization analysis and selecting a larger (smaller) γ_r would thus likely result in yearly results quite similar to those presented for a 30% representational error."

3. *The authors use a fairly simple inorganic PM model in their simulations; it is well known that other inorganic species (such as chloride, sodium and crustals such as magnesium, calcium and potassium) can have an important impact on the aerosol thermodynamics. Could the authors comment the conditions (and perhaps locations) for which the neglected species will have an important impact on the adjoint sensitivity calculations?*

Discussion of the impact of crustal species and sea-salt are expanded upon in Section 4.4, which now reads:

"A possible source of model error is uptake of HNO_3 on mineral dust, which is a source of nitrate aerosol not considered in the model, and thus a possible model bias for locally formed aerosol in the Southwest as well as long-range transport

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of aerosol with dust (Malm et al., 2004; Liao et al., 2007; Fairle et al., 2007). Formation of sulfate aerosol on sea salt is also not included in the model, which can impact sulfate aerosol and HNO_3 concentrations over the ocean and near coastlines (Alexander et al., 2005). However, the largest differences between the observed and modeled nitrate (in the central US) are not likely to be heavily influenced by such interactions."

Further, the impact of other ions not considered in the model on the attainment sensitivities are discussed in the last paragraph of Sect. 5.1:

"While the present work considers only the contribution of inorganic species to $PM_{2.5}$, it is important to keep in mind the role of additional species. When excess NH_3 is present, the sensitivity of nitrate to concentrations of crustal mineral species can be relatively low, at times an order of magnitude less than the sensitivity of nitrate aerosol to NH_3 (Fountoukis et al., 2009). However, in areas where NH_3 levels are lower and mineral concentrations higher, the importance of NH_3 in governing nitrate formation may be diminished. Hence, the non-attainment sensitivities with respect to NH_3 emissions may be exaggerated in the Southwest owing to local dust sources or in the western US owing to transpacific dust transport in the springtime. Still, most of the sensitivities for the present study were located in the central and eastern US."

and later when discussing sensitivity with respect to long-range transport:

"Sensitivity with respect to NO_x emissions may be underestimated as the model does not account for aerosol nitrate associated with transpacific dust transport (Malm et al., 2004; Fairle et al., 2007)."

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