

Interactive comment on “Inverse modelling of CF₄ and NF₃ emissions in East Asia” by Tim Arnold et al.

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This is a paper describing an inverse modelling study for the greenhouse gases CF₄ and NF₃, which are particularly worrisome because of their long lifetimes. These are rarely measured gases with few existing (and quite unreliable) regional emission estimates. Therefore, this study adds important new information on emissions in East Asia. The methods are largely solid and I recommend publication. I have, however, a number of comments that I would like the authors to consider before final publication.

Major

I am somewhat concerned by the large interannual variability of national emissions obtained by the inversion. For example, in Table 1, CF₄ emissions in China in 2012

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are 8.25 Gg/year, but in 2013, they are only 2.82 Gg/year. Is a 65% reduction from one year to the other realistic? This is true also for other species (e.g., NF₃ changes from 1.08 Gg/year to 0.36 Gg/year from 2014 to 2015) and partly also for other countries. For the first example, the change is also outside the combined uncertainty range. I think this needs at least some discussion. How do the authors interpret this? As an inversion artifact? Real changes?

Abstract, line 25: The sentence "Owing to the poor availability of good prior information for this study our results are strongly constrained by the atmospheric measurements." is a wrong statement. The constraint offered by the measurements does not become any better with weak prior information. Of course, relatively speaking, the measurements get more weight in the inversion, but that doesn't mean that the constraint is strong. It also does not mean that uncertainties are lower than with better prior information. In fact, your constraints are very weak for most regions (as you yourself repeatedly point out), which is a consequence of using only data from one station.

The fact that data from only one station were used is problematic in itself. I am aware that there are not many stations measuring CF₄ and NF₃ but was there nothing at all available (e.g., Japanese data)? In my experience, inversions using data from only one site are not very "stable" and the large interannual variability in country-total emissions obtained seems to confirm this.

Line 171+ Lines 183-184: air concentration (dosage), units: The units used for the NAME backward runs don't make sense to me. How can a mass be emitted in backward mode, and how can concentrations be obtained as output? The output should be a sensitivity of receptor concentrations (or, alternatively, mixing ratios) to emission fluxes (either per grid cell, or by square metre, or by volume, possibly by time). But not concentrations.

Equation 3 and text describing it: Why do you include $f_{\text{topography}}$ and f_{inlet} , if they are anyway always 1? For the sake of both brevity and clarity, this should be removed.

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It seems to be something in development that is not yet used. Further, it's a good idea to try quantifying observation/model uncertainty, as this is certainly not a constant (as often assumed in other studies). However, the model uncertainty is not only determined by the boundary layer height at the receptor. Equally important is the boundary layer height in the source regions, and of course there are many other factors determining model uncertainty. Can you discuss/justify/explain your choice of uncertainty scaling, without forgetting to mention that there are many other factors influencing uncertainty?

Paragraph starting at line 318 (and paragraph before): I think it would be good if you could put the scaling factors (posteriors) which you obtain for the MH baseline into a table, or perhaps better even, add the numbers to Figure 1? I assume these are constant values? Or are they allowed to vary with time?

Figure 4: What exactly does the right column show? If I understand right it is the emission flux minus its uncertainty (but I might be wrong, this needs better explanation). But does this quantity make sense? It's the lower estimate of the flux, and all values seem to be negative? Here, I would normally expect something like a map of the uncertainty reduction (e.g., in %) due to the inversion (perhaps does not make so much sense in your case, as prior uncertainties are kind of arbitrary), or a map of the uncertainty itself.

Minor

Lines 44+443: NF3, which contains no carbon, does not contribute to carbon budgets. This needs rephrasing, it's only CO2 equivalents, which you mean.

Line 46: Isn't it misleading to compare such long-lived species to CO2 using GWP100? The lifetime impact of CF4 is orders of magnitude higher than GWP100 suggests.

Line 86: production areas: maybe better production sites?

Lines 200-201: I don't understand what you mean with "the sensitivity of changes in boundary conditions to measured mixing ratios".

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Line 204-205: You need to say which numerical method you have used for cost function minimization. "Non-negative least squares fit" is not concrete enough.

Line 271: What is meant with "If the model boundary layer height transitions across the sample inlet height"? In particular, what is meant with "transitions"? Is it above, or below, or equal, or what? Is there some time development involved, as the word suggests?

Line 276: Equation is not numbered.

Lines 332-334: The sentence "Further, although the very large area for our model domain may not be necessary for this study, the model will not need to be run again should a larger area of analysis be required." does not add any information to this paper. I would suggest removing it.

Line 364-365, "For estimates of emissions from other countries our posterior estimates are not constrained sufficiently to make a meaningful comparison with Fang et al. (2015)." OK, but yet you show the comparison in Figure 3. This is a bit contradictory.

Paragraph starting at line 487: Reference to Figure 7 is missing.

Line 697: "Maps A, D and E". . . D should be C, I think.

Typos, etc.

Line 114: able TO measure

Line 364: Stohl et al. (2015) should be Stohl et al. (2010).

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