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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of volatile organic compound emissions for Europe using data assimilation" *by* M. R. Koohkan et al.

Anonymous Referee #1

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1 Overview

The work by Koohkan et al. seeks to constrain VOC emissions over Europe using EMEP observations and inverse modeling. The content and scope are suitable for ACP. The authors are quite proficient in their application of inverse modeling methods, being well aware of potential pitfalls, and present interesting alternatives to frequently used methods that provide more valuable results. The methodological results related to the statistically consistent non-Gaussian approach are alone of potential wide interest. That being said, the paper would benefit from additional efforts to interpret the significance of the corrections to emissions. Further tests of the reduced model also

appear needed. Lastly, discussion of relevant studies in the literature is lacking. These issues and other more detailed questions are described further below; they should be addressed prior to publication.

2 General comments

- sections 4.2, 4.3. These are great inverse modeling tests, and I just wish the authors could spend a bit more time trying to interpret the results. Does the grouping of errors, or forecast skill, by species indicate anything about the sectors responsible for the errors in VOC emissions? Or the timing (i.e., emissions from a particular season, day of week, or time of day)? Overall the quantitative interrogation of the results is fairly strong; the interpretation is a bit weak.
- I have some issues with the presentation and validation of the reduced model.
  - Figure 2: The log-scale shows the correlation across a wide range of values, which is great, but it does sort of hide the fact that errors on the order of 100% or more abound, for isoprene in particular. Can the authors also present the ratio of the direct to adjoint-based values, or the range of this ratio by species? Do any of the issues with the inversion results for isoprene likely relate to errors in the model estimates on the order of 100%? The authors only really consider lifetime to be an issue, but I think their linearization of the chemistry may also be contributing a lot.
  - The inversion results include changes to the emissions by over an order of magnitude in many locations. Surely this will change the local OH and O_3 concentrations. These would then need to be updated periodically throughout the iterative process, at some frequency determined based on tests (not yet performed) of the extent over which the concentrations in the reduced

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model respond similarly to emissions changes as in the full model. While the authors do test reduced model relative to the forward model around a single atmospheric state, this is not a test of response of the reduced model to changes in emissions, which is necessary.

  - Turner et al (GRL, 2012) consider the adjoint "footprints" of column observations of HCHO. Despite the short lifetime, chemical feedbacks lead to large (hundreds of km) regions of influence. Are such influences for short-lived species missing from the reduced model?

- There are several areas where discussion of the literature is lacking, such as:

  - 33222.20: Discussion of previous efforts to constrain VOC emissions is missing many recent works. What about Stavrakou et al (2009, ACP) constraints on glyoxal? or Liu et al. (2012, doi:10.1029/2012GL051645) with glyoxal constrained aromatic emissions? or Zhang et al. 2011 Atmos. Environ. using HCHO remote sensing to constrain VOC emissions in Texas? These are just a few; the authors should include a more comprehensive survey of the literature, and the statement "have not yet included VOCs" should be removed.
  - 33225.23: earlier, more general works describing the adjoints of chemical transport models exist.
  - 33221.12: The statement "cannot be derived from mass balances... conducted at the source of emissions " is an oversimplification. Many works have used mass-balances to constrain VOC emissions based on ambient measurements. It is indeed a complicated process, but often ratios of different species can be used to generate useful results even for non-conservative tracers. See for example any number of papers where aircraft data or measurements from ships downwind of urban areas are used to constrain VOCs

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during field campaigns. Too many to list here, but I'm sure the authors could find several related to recent campaigns such as CalNex or MEGAPOLI.

  - There has been a lot of inverse modeling work on CH_4, which is a VOC. So perhaps the authors should in some contexts refer to NMVOCs.

- A lot of effort is spent dealing with the fact that direct inversion of emissions can lead to negative emissions, and inversion using L-BFGS-B imposes bounds on quantities assumed to be normally distributed. While it is nice to see the development of case C, where the statistics are adjusted to account for this, it seems like a lot of work compared to a much easier, and common, formulation of this problem: use scaling factors $\alpha = \ln(\frac{e^s}{e^b})$. The emissions are likely to be log normally distributed to begin with (I'm guessing a simple query of the EMEP inventory would demonstrate this to be the case). Further, the authors can use L-BFGS (i.e., not bounded) as this α ranges from minus to positive infinity. At least it would be a nice comparison to B2 and C, hopefully not too much more work.

- The framework assumes 0% error for the boundary conditions. How much might errors in the boundary conditions be impacting the analysis and projecting onto biases in the inferred emissions? What would the authors assume the uncertainty in their boundary conditions actually are, or how much this matters (especially for long-lived species)?

3 Specific comments

- 33221.17: In terms of recent reductions in uncertainties, can the authors provide specific citations?

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- 33223.19: Stable / accurate advection operators are usually nonlinear. Is the one used here really linear? If so, how is transport accuracy affected?
- It is never mentioned what the temporal resolution of the observations is. Hourly? Daily? And at what temporal scales are they used in μ^s? There must be some limitations to the methods used here which rely on the full jacobian H given that d_s adjoint calculations are required.
- 33230.7: I appreciate that calculation of Eq. 15 is computationally intensive, but the phrase "integral over the positive cone" doesn't mean anything to me. Can this be explained more carefully for the lay audience?
- 33227.15: Why is this not expected? I would assume that emissions within particular geo-political boundaries may be correlated, or that emissions for specific sectors would be correlated across broad spatial scales. Also, since the model resolution has yet to be introduced here, it wasn't clear when readying this what might be considered long-range.
- 33231.24: How are boundaries at the top of the model handled? Are these also taken from MOZART 2?
- 33232.5: Could the authors say a bit more here what is meant by approximate adjoint? Is there similarity to the approximate adjoints of Singh and Sandu (2012 Computers and Geosciences)?
- Figure 6: The wording and description of this essential figure is a bit odd. "Normalized" and "correction" may be redundant here. Regardless, I'm not clear if it is showing correction factors, or if showing inversion emissions normalized by EMEP emissions. Further, discussion in the text mentions the large negative source for ISOP in case B1. But from my understanding of this figure, there are also small negative corrections for all inversions for other species, e.g., toluene.

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Interactive Comment

How are we to tell from this figure if these negative corrections are large enough to lead to actual negative emissions? Would that be at the -1 level? Again, clarifying what is actually being shown in this figure would be useful in this regard.

- 332326.8: Exactly! This cannot be said enough, and is unfortunately often omitted from many inverse modeling studies, so I commend the authors here.
- Eq 17: Can the origin of this equation be shown?
- Figure 7: This would have a lot more meaning if the authors included a third column showing the base emissions themselves. Otherwise, it is hard to tell if the relative changes are significant or not.
- Figure 7: For isoprene in France, emissions near the measurement locations show large, localized increases, surrounded by more broad decreases. What is the reason for this?
- 33221.19: How large? 50%? 500%?

4 Editorial comments

- 33220.4: comparison to a standard
- 33221.9: strategies become implemented
- 33222.11: The wording here implies that only the papers discussed after this point used outputs from air quality models as part of the inversion, when such models were as well integral to the satellite-based inversions mentioned at the beginning of the paragraph. Some adjustment to the wording or organization of this paragraph would be useful.

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- I suggest picking one of the names “prior”, “a prior”, “background” or “first guess” and sticking with this throughout the paper.
- 33223.4: the observations and
- 33224.8: Would recommend eliminating “relevant to our study: they are not ”
- 33228.3: It wasn’t clear from reading this paragraph alone how the second solution was obtained. Only later, in discussion of the inverse modeling results, did it become clear that the second approach is a gradient-based iterative minimization of L_s using L-BFGS-B. It would be nice to revise / expand this paragraph to make the method clear here.
- 33232.22: Can the phrase “value screening” be clarified?
- 33237.11: One way to understand this is that . . .

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 33219, 2012.