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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Multi-species inversion and IAGOS airborne data for a better constraint of continental scale fluxes" *by* Fabio Boschetti et al.

Anonymous Referee #3

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General comments:

The paper presents a multi-species inversion framework tested using pseudo-data experiments. Various assumptions are made to evaluate the sensitivity of the inversion, with an emphasis on the impact of error correlations across species and sectors. Overall, the paper presents an innovative approach to assimilate various atmospheric species in a single inversion framework. This study is clearly worthwhile publishing but lacks a better evaluation of the aggregation operator assumption (perfect prior emission distribution) and the impact of systematic errors in the system affecting the correlations in the gas-sector attribution problem. The Observing Ssystem Simulation Experiments (OSSE's) cover some of the assumptions with varying levels of uncertainties but several components are not carefully considered. The two major concerns here are the

aggregation operator, that remains perfectly known and so the spatial distribution of the prior fluxes, and the assessment of correlations among sectors and across trace gases for the different species that remain very unclear. A last but less critical concern is related to the assumption that transport errors are similar across species, which is unlikely for CH_4 and CO_2 for example, rarely co-emitted (only CO_2-CO is discussed) and therefore affected by different problems in different parts of the domain. The work focuses primarily on random errors and ignores systematic errors that remain the main limitations in atmospheric inversions. Therefore, this study requires some additional experiments before publication, specifically addressing the error associated with the aggregation operator and errors in gas ratios for the different sectors.

- The use of an aggregation operator needs to be discussed. Hyper-parameters (here scaling factors for the sectors) are used to reduce the dimension of the problem but corresponds to an assumption of perfectly-known distributions. The system should be evaluated not only under the "perfect spatial distribution" assumption, especially for CO_2 biogenic fluxes which are clearly not well-known. One suggestion to clarify the concern here would be to use VPRM as truth but assumes a different distribution when constructing the aggregated solution such as the posterior fluxes from Panagiotis et al. (2016). Other experiments could be designed here to test the aggregation problem. Similarly, the area defined by half of the total footprint is arbitrary and never tested nor justified. Why 50% was used? How much variations are expected within that area which would affect the error correlations? If a power plant is located near an airport, how would that affect the CO/CO_2 correlations and therefore the homogeneity within the aggregated area?

- The discussion about error correlations across species is confusing. How did you define the emissions for the different sectors? Have you assigned gas ratios to various sectors? If so, what are these ratios? Some of the discussions are related to using CO_2 and CO data to diagnose gas-to-gas correlations, but the exact definition of the emissions of the different gases for each sector has been defined in the inversion system. Or maybe the sectors are unrelated for each gas? The different sectors have ratios int erms of trace gas emissions but these emission ratios vary regionally. This section needs to be explained in more details. The assumptions made here should also be tested in the inversion framework.

- CO biogenic fluxes: the paper does not address the problem of CO biogenic fluxes during the growing season. Warm days in summer correspond to large amount of biogenic VOC's beign emitted from the vegetation, producing CO to non-negligible levels. This issue should be discussed if not addressed. How would this problem affect the ability to retrieve the truth?

- When you constructed your error correlations for CH4, transport errors are unlikely to be highly correlated as CH4 is only partially co-emitted with CO2 and CO. Large emissions from NG production and farming activities are uncorrelated with biogenic or fossil fuel consumption. This problem should be adressed here. If transport errors, which are spatially variable, affect CH4 and CO2/CO in different ways, the error correlation would be affected. Additional experiments using incorrect error correlations would quantify the sensitivity of the inverse fluxes to the assumptions made in prior errors.

- The problem of unreported sources in CH4 inventory is not addressed at all. Recent papers have discussed the lack of information for natural gas and oil production operations, or from recent and old mining areas. How would unreported sources affect the inverse solutions? This question comes back to the aggregation operator.

- The utility of the figures showing the multiple error covariance matrices for the different cases remains limited. The information content would be better described with words or mathematically. Readers cannot extract useful information from contour plots of covariance matrices. They could remain part of the paper but as part of the supplementary information. A table could also synthesize the various assumptions tested in the inversion system.

Technical comments:

3-1: Consequently, intercomparisons...

3-3: the international level

3- 1st paragraph: This paragraph is confusing and not always following a logical path. Prediction skills and emission reduction are two different problems not directly connected to each other. Explain better the broad context of this study by focusing on the main general issues and clarify which one you are trying to address here.

3-10: A commonly used approach to estimate...

3-13: Actually, the uncertainty reduction relies purely on the assumptions made in the system and not on the effective ability of the system to produce a reliable solution. Bayesian system assumes that data will improve the a priori by construction. Explain better what you mean here.

3- 2nd paragraph: Several papers are missing here. For example, CO2-CH4 inversion using satellite data (Pandley et al., 2015) or the optimization of co-emitted species (Brioude et al., 2012), and early work on delta 13-CO2 by Enting et al. (1995). THe authors should dig into atmospheric chemistry studies where several studies have addressed the use of multiple co-emitted species to constrain emissions at small scales.

Previous studies using multiple species to constrain emissions should be introduced here, even without having used a formal inversion framework, such as urban studies over Los Angeles (e.g. Peischl et al., 2013). The optimization problem is equivalent and relies on similar ideas to constrain the emissions.

5-24: This technique assumpes that the wind direction and speed are comparable near the surface and at 2km high. Mass-balance studies have shown that this is often not the case (e.g. Karion et al., 2015). Free troposheric air represents different air masses due to the wind direction and speed gradients in the vertical. This assumption would need to be tested with the particle model.

7-3: What about CO biogenic fluxes? During warm summer times, biogenic CO fluxes represent a significant fraction of the signals. Did you ignore this contribution in your study?

2.1.3 To reduce the dimension of the state vector, you assume here that the spatial distribution of the prior fluxes and emissions are perfect, using an aggregation operator. This approach is reasonable for fossil fuel emissions but less convincing for biogenic fluxes.

12-21: How did you take into acount the truncation of the prior errors? Did you adjust the truncated random perturbations to match the non-truncated assumption made in the prior error covariance matrix? 14-13: The expression "lion's share" should be avoided. A fraction of the contribution is a better metric to describe the importance of the sector.

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