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Comment

Interactive comment on “The formaldehyde budget as seen by a global-scale multi-constraint and multi-species inversion system” by A. Fortems-Cheiney et al.

Anonymous Referee #1

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Fortems-Cheiney and co-authors apply a multi-species inversion analysis with a 3D model to simultaneously constrain a 1) surface emissions of CH₄, CO, and methyl chloroform (MCF) at 3.75x2.5 degree resolution; 2) CO, CH₄ and MCF initial conditions (8-day, 3.75x2.5 degree resolution); 3) scale factors for 3-D production of HCHO from VOC oxidation (8-day, 3.75x2.5 degree resolution); and 4) scale factors for OH concentrations at 8-day resolution in 4 latitude bands. The analysis employs satellite observations from MOPITT and OMI (CO and HCHO) and surface observations (CH₄, MCF) as the observational constraints in the inversion. They infer various changes to above 4 sets of variables on the basis of their analysis, and interpret those changes (briefly) in terms of the underlying drivers.

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Discussion Paper



The analysis is novel in optimizing for these variables simultaneously, and in the way that it incorporates a range of space-based and in situ observational constraints to get at the questions of interest. It is also smart in the way that it exploits the interrelationship of the CH₄-HCHO-CO system. The topic is certainly appropriate to ACP. The main weakness that I see is that it doesn't do a sufficient job of exploring and demonstrating the extent and resolution of the constraints that this observation and analysis system can really provide. I have some specific comments below that I feel should be addressed before this is published.

————— Scientific comments —————

- I wonder how well we can expect the inversion to independently resolve all these different variables that are being optimized simultaneously. Clearly many of them are strongly coupled. For instance, how well can a bias in model OH be resolved from a bias in the surface fluxes of CO/CH₄/MCF? I feel the authors need to demonstrate the robustness of their approach for us to give the findings full credence. For example, one could do this by generating pseudo-observations (with errors applied) with different aspects of the state vector perturbed, and test how well the inversion can get back to the true solution. And without messing up some other aspect of the state vector! Perhaps I'm wrong, but it seems likely there is some degree of conflation between those sets of variables being optimized.

- I guess much of the constraint on OH is coming from MCF, especially since the assumed error on MCF emissions is only 1%. This 1% is not a physically reasonable assumption. Current MCF emissions are fairly small, but they are probably mostly from banks, stockpiles, landfills and so forth at this point, and so quite uncertain in a relative sense. For example, US emissions during 2003-2006 were shown by Hurst et al. (JGR, 2006) and others to be ~60x larger than EPA estimates. How does your solution change if a more realistic uncertainty on MCF emissions is used? I'd be hard-pressed to think of any compound whose emissions are known to better than 1%! Especially on a grid-basis, as you're doing. We likely have a reasonable constraint on the global flux

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(though certainly not close to 1%), but much less so on a grid-cell by grid-cell basis.

- please discuss how uncertainty in model OH in high-isoprene, low-NO_x environments might affect your solution. MCF provides a constraint on OH, but only in a mean sense as it has a 5-year lifetime. You could well have an OH bias that, in the tropics correlates spatially to a degree with one of the variables you are trying to solve for, P(HCHO).

- what does it mean to be optimizing OH separately from and independently of P(HCHO), which in reality is driven by OH?

- Since your model includes the effect of OH on CO and CH₄, but not vice-versa, is there potential for a result that is not self-consistent? In other words, CO and CH₄ are the main global sinks of OH, but to my understanding this dynamic isn't captured in your model framework, only the effect of OH on CO and CH₄.

- to their credit, the authors carry out a bootstrap analysis to put some error estimates on some aspects of their solution. But they do not assess how their solution might change if different errors are assumed for the prior and for the observation system.

- model transport is another important source of potential error which is not addressed or discussed.

- The GEIA isoprene emissions are now more than 17 years old, and the following version, MEGAN, has been available for more than 6 years. I wouldn't insist that the authors switch to MEGAN, but this does seem odd and in my view somewhat reduces the impact that their findings could otherwise have.

- For the simplified (5-reaction) chemistry being employed, which assumes methane to formaldehyde in a single step, one thing that is neglected is the NO_x dependence. For example at low NO_x you're more liable to form methyl hydroperoxide, which would delay the HCHO production by a couple of days, and slightly reduce it to the extent that CH₃OOH is deposited. I guess this would be a minor effect but worth mentioning.

6921 and Fig 4. Why does the agreement get worse over W. and E. Europe?

6923, when attributing changes in P(HCHO) as a result of the inversion to VOC emissions, how do you know it's not the HCHO yield that's wrong? Can you separate the two? Particularly in tropical or low-NO_x areas, how do you know the timescale and/or yield for HCHO production in the model is accurate?

6927, how do you reconcile this CO emission difference with the Kopacz and Hudman papers? Hudman et al. showed that their source estimate was consistent with aircraft observations. Given that, I don't see how a factor of 3 increase compared to their value is physically reasonable. "it can be due to an underestimate of the CO traffic emissions in the EDGAR-v3.2 inventory"? That could explain the change from your prior to your posteriori, but not the discrepancy between your posteriori and the other studies.

6930, 10-11 "we demonstrated the robustness of our multi-species inversion system", not sufficiently, see comments above.

- 6917, is Fung et al. (1991) really the most current estimate for this?

————— Technical comments —————

6910, opening paragraph of introduction. By talking about the direct HCHO sources first, you give the impression that these are most important when in fact they are quite small.

6911, 4-5 and 17: "HCHO modulates the budget of CH₄, CO, and NMVOCs", and "HCHO and its interactions with CH₄ and NMVOCs". Ok for CO, but for CH₄ and NMVOCs this is odd phrasing, since HCHO is produced from them rather than the other way around. HCHO doesn't really modulate the budget of CH₄ and NMVOCs, or interact with them, at least in any direct way. Did you mean indirectly as a source of HO_x?

6911, 8 (and also a similar comment at the beginning of 5.2.1): "NMVOCs and their short lifetimes". In fact lifetimes vary from hours or less to weeks or longer.

6912, 8: not sure I'd agree that there exists a "consistent global measurement network"

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for NO_x. Unless you mean NO₂ from space, but that isn't NO_x.

6912, 10: “these limitations can now be addressed by using HCHO total columns retrieved by satellite”. Careful not to make it sound like measurement errors are now a thing of the past – the satellite data of course have their own errors and uncertainty!

6912, 18-19: “However, none of these studies combine retrievals for various species together”. That's not a true statement. What about Stavrakou et al. (ACP, 2009) who inverted simultaneously for HCHO and CHOCHO? Also, Barkley et al. (JGR, 2008) and Marais et al. (ACPD, 2012) both combined space-borne HCHO and NO₂ measurements, though NO₂ was used as a diagnostic for fires and chemical regime rather than as something to be optimized.

- 6914, 4: On the ACPD page for the Kim et al. paper, it says “publication in ACP not foreseen”. Nothing against that paper in particular, but what is the protocol for citing papers that did not make it through peer review? Do we want to be doing that?

- 6914, 7: since TPK is a co-author it seems unnecessary to cite a personal communication in this way.

- 6915, 23: Also a minor ocean sink for MCF, cf Wennberg et al. (GRL, 2004).

- 6925, 5-8, unclear, awkward.

- Section 5.2.3 – Section on changes to the global HCHO sink. Doesn't the change in the sink just come about as an inevitable consequence of the change in the source? Doesn't seem to need its own section.

- section 5.3.1, “copares”, “fulle”

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