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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "First space-based derivation of the global atmospheric methanol emission fluxes" *by* T. Stavrakou et al.

**T. Stavrakou et al.**

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We would like to thank the reviewer for his/her thoughtful comments. In the following, we address the concerns raised. Reviewer's comments are *italicized*.

**General comments**

*- my main complaint with the paper is the lack of any real uncertainty analysis. The authors discuss the uncertainty in the IASI measurements themselves but do not discuss in any real way the uncertainty in their inversion results. I see this as problematic and needing to be addressed before the paper can be published. Otherwise one doesn't know how to interpret the results. For instance, the a posteriori biogenic flux is 100*

*Tg : : : is this 100.00 +/- 0.01? +/- 100%? 200%? I recognize it is not necessarily straightforward to come up with realistic quantitative uncertainty estimates on inversion results and that the comparison with independent observations is one step in this direction. However I think it is incumbent on the authors to do a bit more in this regard. For instance, how do the retrieved fluxes depend on uncertainties in the overall model framework? E.g., model OH concentrations? Meteorological fields? Assumptions regarding reactive uptake during dry deposition (Karl et al., Science 2010)? Etc? Without treating these points the paper risks becoming another inversion study lacking a good-faith assessment of the robustness of the results. It deserves better!*

To address this criticism, two new sections (9 and 10) have been added in the revised manuscript.

Section 9 deals with the a posteriori error estimation of the derived fluxes. To determine the a posteriori error covariance matrix, we used an off-line iterative approximation of the inverse Hessian matrix based on the Davidon-Fletcher-Powell updating formula. The achieved error reductions (ratio of a priori on a posteriori error) are more important for the biogenic emission source, and reach 3 over the Former Soviet Union, 2 over South America, 1.64 over Europe and 1.54 over Northern America. Error reductions are calculated also for the vegetation fire source, but they are less significant.

Besides the uncertainties on the a posteriori emission fluxes, in Section 10 we propose a tentative assessment of model errors, based on a set of sensitivity inversions conducted to investigate the influence of uncertainties on meteorological parameters, on methanol sink processes, and on the a priori methanol plant emission source. Although this evaluation is not exhaustive, it accounts for the most important uncertainties associated to the model. Table 5 summarizes the performed inversions and provides the corresponding tropical (25 S-25 N), extratropical and global biogenic source inferred in each case study, and Figures 13-15 illustrate the changes derived from the sensitivity inversions. The inferred emission estimates are found to be quite robust in the different sensitivity cases, with global estimates differing by less than 10%, although



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differences on the order of 30% are found on the regional scale (see Sect. 10 for a detailed discussion).

*- One point that is not discussed: how well do you expect the retrieval to perform over fire scenes?*

There are several aspects. The largest methanol concentrations are usually observed in local fire plumes (see Coheur et al., ACP, 2009). If the plume ascends to the free troposphere, the absorption will be significant and we expect the retrieval algorithm to perform well. However, the retrieved column will be conditioned by the shape of the a priori profile, which might not always be appropriate. The resulting errors are hard to quantify and will depend from case to case. The presence of aerosols can also increase the retrieval errors.

*- And a related point: to what extent do you really have constraints on the biomass burning flux, given that it is only 5% of the biogenic flux? This gets back to the a posteriori uncertainties.*

In the new section 9 on the a posteriori uncertainties on the derived emission estimates, we provide the error reductions for the vegetation fire source : on the order of 14% on the global scale, 10-12% over Africa and southern Asia, and almost negligible error reductions over other regions. These error reductions are modest because the biomass burning source represents only 3% of the global methanol source.

*- How well are the a posteriori fluxes (biomass burning versus biogenic) really resolved? This is not assessed in the paper. Figure 4 seems to imply some conflation of the two source types. For instance, fires in Alaska and E. Russia appear to be resulting in artificially higher biogenic fluxes in those areas.*

We acknowledge that some conflation of the two sources might exist. However, at most locations/times, one of the two sources is largely dominant, and is therefore well



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constrained by the observations. The simultaneous optimization of two source categories is also facilitated by the prescribed spatiotemporal correlations of the a priori errors ($B$ matrix in Eq.11), which tend to preserve the spatiotemporal patterns of the a priori emissions in each category. For example, over Alaska, large fires occurred in July/August 2009, resulting in methanol emissions about an order of magnitude larger than the biogenic emissions in that region. Conversely, the biogenic emission increases seen in Fig. 4 over Alaska and Yukon are mostly due to a strong enhancement in May-June, when biomass burning emissions are negligible. We have added a comment on this in Section 8 of the revised manuscript.

*- since you are not doing full retrievals everywhere, is there potential for differential bias depending on the surface properties and hence land cover of the retrieval scene? And then that such bias is erroneously attributed to emission differences? For instance, you discuss specifically an apparent model underestimate in arid regions. The model underestimation in the free-troposphere is touched upon in multiple places, and the strength of the oceanic source/sink is given as a possible cause. A bit more more could be said on this point. For instance, would a stronger ocean source (sufficient to resolve the discrepancy in the free troposphere) give a realistic simulated vertical profile over oceans?*

Biases related to surface properties should be low, since we have performed retrievals on a representative set of scenes with varying surface properties (see Razavi et al., 2011). In fact, land cover is not such an issue; the retrievals are more conditioned by the temperature gradients in the troposphere.

Regarding the model underestimation in the free troposphere, the results of two additional sensitivity runs using either doubled or quadrupled oceanic emissions of methanol indicate only a minor influence of oceanic emissions on free tropospheric mixing ratios over the INTEX-A campaign area : the mean model mixing ratio between 4 and 11 km altitude is increased by 3% (to 0.983 ppbv) when the oceanic source



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is doubled and by 11% when the ocean source is increased fourfold (to 1.056 ppbv), but it stays well below the observation (1.65 ppbv). A better agreement with the observed free tropospheric mixing ratios is achieved when an enhanced vertical transport by cloud convection is considered, or when prescribed climatological OH fields are used (mean model vmr: 1.125 ppbv and 1.163 ppbv, respectively, cf. Sect. 10 and Fig. 14). Convection might be currently underestimated in the model and/or OH simulated concentrations could be overestimated in this region. Alternatively, the missing upper tropospheric methanol in the Eastern US could be at least partly due to eastward transport from the Western US, where the modelled columns are still underestimated after optimization (Figs. 3 and 4 of the revised manuscript). These points are now briefly discussed in Section 8.

**Specific comments**

*- 5223, 15-16: use consistent units to avoid confusion (Tg or TgC)*

Changed

*- font on some figures is too small; hard to read*

Fonts have been enlarged in Figs. 1, 2, 3, 4, and 7 of the revised version.

*- Figure 5, 6, and especially 7, suggest a different color and / or symbol for the aircraft observations and the S1 simulation, it can be hard to distinguish the two (for instance, the middle panel at the lowest vertical level)*

Done

*- Section 8, page 25, paragraph 3 (and elsewhere): What exactly is meant by "satisfactory agreement"?*



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Reworded

*- Figure 11: Add in IASI error bars here for consistency*

Done

*- Section 7, page 21: You mention here that the seasonal cycle of IASI in the SW US is similar to that observed at Kitt Peak, but neither the western nor SW US are shown in Figures 5 or 6.*

We have now included a new figure showing a comparison between FTIR, IASI, and model methanol columns before and after inversion (Fig. 12). The figure illustrates the good consistency between IASI and FTIR measurements in this region. When constrained by IASI measurements, a better model/FTIR match is achieved, despite persistent underestimations. The correlation coefficient between the model and the FTIR data is equal to 0.55 in S1, 0.8 in S2, and 0.94 after inversion.

*- Section 7, last line, reference to Figure 9: Statement is confusing b/c satellite data is not plotted in Figure 9. Do you mean that the OptS2 simulation results in better agreement with the surface observations, thus the satellite data must be consistent with those observations? Please clarify text accordingly.*

The statement has been changed in the revised version : “The OptS2 simulation results in better agreement with the surface observations at different European sites, as depicted on Fig. 9, implying consistency between satellite observations and ground-based measurements."

*- S240, 25: Please be more specific in your discussion regarding Indonesia. Are you referring specifically to the Sumatra plot when you cite the 2x discrepancy over Indonesia? But then you discuss in-situ observations over Malaysia, which means we should be looking at the Borneo plot? Indochina technically should not include either*



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*Indonesia or Malaysia, but readers might easily assume you mean to refer to the Indochina plot, which actually shows good agreement for the May-June timeframe (when the in-situ data were taken).*

The Borneo plot of Fig. 5 comprises a much larger region (6S-6N, 110-120 E), whereas the OP3 campaign site is located in northeastern Borneo, at approximately 5 N, 118 E. The comparison we mention in the manuscript concerns the model grid cell which comprises the OP3 site. This is now made clear in the revised manuscript: “the S2 methanol concentrations calculated at the model grid cell comprising the campaign site located in northeastern Borneo (5 N, 118 E) are by 2.5 times too high..."



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