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Atmospheric Chemistry and Physics Discussions

Interactive comment on "First global distributions of methanol and formic acid retrieved from the IASI/MetOp thermal infrared sounder" *by* A. Razavi et al.

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Reply to Referee #3

Overall assessment.

This paper provides a first 1-year global data set of methanol and formic acid columns from the IASI instrument. It is an important contribution to atmospheric chemistry that will help to better constrain the atmospheric budgets of these two species. I recommend publication after consideration by the authors of the specific comments

below. Although the paper is overall well written and of high scholarly quality, there are a number of simple grammatical mistakes in the text that I did not itemize here but that the authors can pick up with a careful read. Also the labels on several of the figures need to be enlarged.
We thank the referee for his comments. Correlations in section 5 were removed and replaced with the comparison of methanol, formic acid and carbon monoxide time series as also suggested by the other referees. Individual replies for each comment are given below. The text has also been corrected for grammatical mistakes and some labels were enlarged for clarity.

Specific comments:

1. Page 21476, line 3: the second sentence of the abstract is misleading (cf. comment 2), unnecessary, and does not stem from the work presented here. I suggest cutting.
The sentence has been removed.

2. Page 21478, line 27: The effect of methanol on OH is only a few % (cf. Tie et al. cited in text). I wouldn't call it "significant", perhaps "minor" instead.
"significant" has been replaced by "noticeable".

3. Page 21479, line 19: I am not familiar with this post-sunset "boundary layer squeeze" effect in the methanol data. If it is indeed a common feature then I stand corrected but surprised - the biogenic methanol source is thought to shut down at dark.
We agree with the referee that this feature observed by (Nguyen et al., 2001) is uncommon. The sentence has therefore been corrected :
"The concentrations usually increase during daytime due to the light induced release

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of methanol by plants."

4. Page 21485, line 8 and Figure 2: it's not clear to me what these covariance matrices actually represent.
These covariance matrices represent the variability of the a priori profiles calculated from simulated profiles of the IMAGESv2 model. These matrices are introduced in the optimal estimation method to account for the correlation between the different altitude levels and their possible range of value. This has been clarified:
"These two profiles are illustrated on Fig. 2 together with their associated covariance matrix (which represent the model variability for the profile -diagonal elements-, and the correlation between values at different altitudes -off diagonal elements-)."

5. Page 21489, line 7: would the higher surface T from fire also cause some error in the retrieval method?
Higher temperatures at the surface may provide higher sensitivity of IASI towards the lowest layers. This has been included in the following sentence :
"Although biomass burning is assumed to be a weak emission source of methanol (accounting for less than 5% of total emissions according to current inventories) (Jacob et al., 2005), the main hot spots in the global distributions appear to be possibly related to fires. This could be caused by the fact that methanol emitted by fire events is usually transported higher in the troposphere where the IASI sensitivity is larger or by the fact that a better sensitivity near the surface is induced because of higher surface temperatures for burning areas."

6. Page 21490, line 5: To what extent can the error in the methanol retrieval be considered random vs. systematic, i.e., reducible through a large number of observations? Same question for formic acid.
On a global yearly scale, we do not expect systematic errors, because the regression

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coefficients were determined on a representative set of atmospheric conditions. Systematic errors cannot be excluded in particular circumstances such as in the presence of extreme values of ozone or water vapor. We have clarified this in the text.

7. Page 21493, line 6: anthropogenic sources of formic acid would be presumably aseasonal and this is not apparent either over the eastern US or eastern China. In the case of the eastern US, the seasonality suggests a biogenic source. In the case of eastern China, the seasonality suggests a source from burning of agricultural waste (see Fu et al. 2007, cited in paper, who find the same enhancement for HCHO).
We thank the referee for this useful comment. Indeed, the formic acid total columns in eastern China do not seem to experience a strong seasonal variation from MAM to SON 2009, indicating the possible contribution of anthropogenic activities. The total column above eastern China during DJF is unfortunately not available because the thermal contrast is lower than 5 K.
For eastern USA, only the MAM and JJA seasons are available but we agree that the increased total columns during JJA may be related to biogenic emission. This has been corrected.

8. Page 21493, line 14: I'm not getting anything from this whole Correlations section. The correlations could simply reflect similar but independent spatial distributions. I suggest reserving this section for a future paper comparing model to observations, where the observed correlations would provide a useful test of the model.
Section 5 has been modified and includes now the figure 1 (in the pdf attachment) which compares time series of methanol, formic acid and carbon monoxide in biomass burning regions. A fair correlation between these three species is observed in Brazil, Congo and SE Asia with a certain time-lag, however. Although biomass burning is the likely source, it is obvious that more work will be needed in the future to identify/quantify sources by sector. Work in this direction, using the IMAGESv2 model, is ongoing.

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Section 5 reads now :
"In this section we compare the 2009 time series of methanol, formic acid and carbon monoxide for three selected regions subject to biomass burning. The comparisons are most likely to reflect similarities/differences in the free tropospheric columns, as IASI is more sensitive to this altitude range for these three species. Monthly mean total columns of methanol and formic acid are shown in Fig. 11 together with the total columns of carbon monoxide and the AATSR fire counts for three $10 \times 10^{\circ}$ regions located in Brazil (15–5°S, 60–50°W), Congo (15–5°S, 20–30°E) and South-East Asia (20–30°N, 95–105°E). For each regions, the time series of CO, CH_3OH and $HCOOH$ are similar. An increase in the total columns is observed for the three species just after the month with the maximum fire counts. The highest number of fires (exceeding 700) is found above Congo where methanol, formic acid and carbon monoxide reach high values, with increases of about $1.6\,10^{16}$, $2.5\,10^{16}$ and $1.4\,10^{18}\,molec/cm^2$ in comparison with their mean total column between January and June, respectively. In each cases, the CH_3OH maximum lasts longer than for CO or $HCOOH$ suggesting an additional source. Overall higher concentrations of CH_3OH found above Brazil and Congo might be due to the larger biogenic source in these regions.

In addition to looking into correlations regionally, preliminary global analyses were carried out. Linear correlations ($R^2 = 0.7$) between CH_3OH and $HCOOH$ were found during the DJF and SON periods highlighting specific emissions or fate of these two species."

The conclusion section has also been modified accordingly :
"Time series of methanol, formic acid, carbon monoxide and AATSR fire counts were also compared and found to be fairly well correlated for three different regions (Congo, Brazil and South-East Asia) where biomass burning is their likely common source."

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