

## ***Interactive comment on “IASI measurements of reactive trace species in biomass burning plumes” by P.-F. Coheur et al.***

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Received and published: 10 July 2009

Unfortunately, one of the reviewers for this paper after providing an initial positive quick-review has not delivered a review for this paper. Therefore, I have listed a few comments and suggestions of my own below in spite of not being an expert in IR remote sensing.

The manuscript reports on the detection of the signature of reactive trace gases in measurements of IASI during biomass burning events in Siberia and Greece. Examples of the spectral identification, profiles retrieved and plume evolution are shown and discussed. In addition, first applications of the IASI results for estimating total emissions and relative lifetimes are presented. The paper is well written and reports on exciting new satellite measurements with large potential for future applications. It is

C2618

rather technical and would have fitted better into AMT than ACP. In the context of a special issue on IASI first results, I think that is also acceptable for ACP. However, as already noted by the other reviewer, the paper does unfortunately not provide discussion on the uncertainties, vertical sensitivity and assumptions made in the retrieval. In addition, some of the claims made are not well supported. I therefore can only accept the paper after major revisions as suggested below and requested by the other reviewer.

R: We thank the editor for his detailed review and his enthusiasm on the capabilities of IASI to measure new reactive species of the Earth's troposphere. We certainly understand his concerns on the lack of detailed error budget, posterior characterization etc. . . , which were also raised by the referee. We have added a series of relevant information to address these concerns and have updated the Figures as suggested. We hope this would answer the principal issues and that the paper be acceptable in its revised version for publication in ACP. Our detailed responses are provided below.

Major comments: \* My main concern with the paper is that it does not provide any idea on the uncertainties in the numbers given. No errors are discussed, no comparison is made to independent measurements, no averaging kernels or weighting functions are shown. Also, possible effects of aerosols and clouds which are certainly present in part of the fire plumes discussed are not mentioned at all. Even though no complete error discussion might be necessary for a paper showing very first results, basic discussion of uncertainties cannot be omitted.

R: As explained also to the other referee, the retrievals have not been performed focusing on vertical profiles, but more on the detection and the first coherent estimates of concentrations in the plumes, which are needed to support the analyses as a function of space and time. The paper provides the first evidence that this is possible with IASI and these unexpected results open in our opinion promises for future dedicated researches.

C2619

Considering this initial objective, there have been no attempts here to use appropriate and sophisticated prior information. Especially the a priori covariance matrix assumed is a simple ad hoc variance-covariance matrix with extremely large variability and unrealistic correlations (7 km with an exponential decay). The large variability was needed to capture the enhanced signals in the plumes, starting from background prior profiles (from standard atmospheres -NH<sub>3</sub>- or global annual averages from recent chemistry models -C<sub>2</sub>H<sub>4</sub>, CH<sub>3</sub>OH-). For these reasons, the posterior characterization in terms of errors and averaging leads to mostly unrealistic results.

However we certainly agree that the elementary aspects of retrievals and of the errors were missing in the manuscript. The following changes have been made:

- A full paragraph giving the details of the retrievals has been added (end of section 2.3.). It includes a description of the a priori profiles and of the prior ad hoc variability assumed.

- The statistical errors on the retrievals have been added in the text for each column given, insisting, however, that these are likely unrepresentative and underestimated because of the assumption on the prior profile and the variance-covariance matrix. In order to test this we have compared a few of the NH<sub>3</sub> columns from this work with those obtained starting from a more appropriate set of retrieval parameters recently built (paper in preparation by Clarisse et al. The reference to it has been added where appropriate). We come to the conclusion that the columns could be affected by the choice of the prior by more than 30%. This remains, however, in the range of the statistical errors provided. Other sources of errors may arise from e.g. the plausible presence of aerosols in the plume. these have not been taken into account.

- The DOFS are around 1 for all species –this is added in the text-, showing that only a column or sub-column would be meaningful. The averaging kernels are not shown, for the reasons exposed above. It should be mentioned here again that it was not our intention to suggest that vertically resolved profiles could be retrieved for these

C2620

species (hence the few details on this) and we apologize if the reader was left with this impression. The profiles shown in Figure 5 are of course fully indicative, mainly corresponding to a scaled prior profile, and this has been added in the text: “with volume mixing ratio near the surface (essentially scaled versions of the prior profile due to the absence of vertical information) . . .”

\* In many respects, the reader is left without information on what exactly was done. For example, it is not clear which a priori profiles were used for the different gases. In Fig. 5, the vertical distributions appear very different for the three species and it is not clear to me, why that should be the case if they are all products from the same fire. As already mentioned by the other reviewer, the interpretation of the profiles is also not clear as nothing is said on how many pieces of information the measurements shown provide in the troposphere. I assume that the information is mainly on the column and not the vertical distribution and that the profiles shown actually are the scaled a priori profiles.

R: See above: Only for NH<sub>3</sub> there is some vertical information, but not enough to provide (DOFS close to 1.3) meaningful vertical profiles.

\* The central part of the paper is the demonstration that IASI nadir measurements can actually be used to detect the reactive species discussed. This is done by comparing the difference of measured and modelled spectra where one of the species has been omitted with the spectral signature of this species. While the results are very convincing for NH<sub>3</sub> and acceptable for C<sub>2</sub>H<sub>4</sub>, I don't think that the results for C<sub>3</sub>OH are clear, neither in Fig. 3, nor in Fig. 5. Also, in Fig. 4, the only obvious signal is HCOOH while all the other structures discussed are uncomfortably close to the noise level. While this does not imply that the measurements do not contain information on the respective species, it is by far not as clear as it appears in the text and without discussion of the uncertainties, I don't think one can proceed to the interpretation of these results.

R: The Figures have been fully revised to better reveal the spectral signatures of the

C2621

different species. This has been done in Figure 3 and in Figure 5 (see Figure 1 attached) by plotting the residuals on a scale per species and in Figure 4, by adding the residual of the 'best fit', taking all the species into account. In these new Figures, the Q branch of CH<sub>3</sub>OH appears better (it remains close to the RMS; but the latter is in fact 2-3 times larger than the instrumental noise in that spectral region). Especially in Figure 5 the signature is clear, with the entire P, Q, R band structure that is seen. In recent plume analyses, the signature was found to be even stronger, detectable on the raw spectrum despite the overlap by ozone. In Figure 4, the detection to HCOOH is indeed unambiguous. We have revised the wording throughout the manuscript to be more careful for the assignment to PAN, although the spectral feature at 780 cm<sup>-1</sup> is in our opinion well matching the expected signature. The low errors on the column retrieval of that species (the errors have been added in the text) tend to confirm this. The assignment made to CH<sub>3</sub>COOH is more tentative but this is adequately said in the paper.

\* An important issue for interpretation of satellite data is not only the detection of enhanced signals but also the noise level of measurements where no signal is expected. I therefore think that all IASI measurements of the respective orbits should be shown in Fig. 7 and Fig. 6, demonstrating how close the values are to 0 outside of the plume

R: The Figures 2 (see Figure 2 attached) and 7 have been revised to include also the noise levels. In Figure 7, which consists in an addition of different IASI overpasses, only a subset of points outside the plumes was considered to avoid overlaps; still the Figure now clearly highlights the enhancement in the plumes as compared to the background and we thank the referee for suggesting this correction. Figure 6 was not changed because for this purpose only the points showing significant signatures have been analyzed (thus already filtered out for noisy values); this has been clarified in the Figure caption.

\* One point made several times in the manuscript is that the results show that IASI has sensitivity down to the boundary layer. While I agree that IASI has sensitivity to the

C2622

lower troposphere under favourable conditions, I don't see how this was demonstrated in this paper apart from the rather vague discussion of probable plume height. If this claim is made, it needs to be substantiated either by showing the vertical sensitivity of the measurements from radiative transfer calculations or by validation with independent measurements.

R: The editor is right to pinpoint to the absence of enough supporting evidence for these statements. The dependency of the spectra to the temperature profile, in conjunction with the fact that the species are "likely" confined in the boundary layer (because of their short lifetime), would indicate that IASI probes down to the surface. However, this indeed comes in assuming that the fires did not eject the plume at higher altitude and we don't have sufficient proofs for this (in particular there are no independent measurements that we can use, except for the Greece fires, for which Calipso measured a low altitude aerosol layer, consistent with the IASI observation). Basically the spectrum in emission is probably the best proof of IASI sounding the boundary layer as it can only be explained by the presence of tropospheric layer hotter than the surface, which is picked up in the L2 data 1km above the surface. This could be demonstrated by a series of forward transfer simulations and could also be supported by appropriate profile retrievals (using adequate variance-covariance matrices) but this would then require substantial technical additions in the paper, which we think are outside the scope of this work and would not fit for the special issue. For these reasons, we have chosen to change the text throughout the manuscript in such a way as to avoid strong statements, referring instead to a paper by L. Clarisse in preparation, where all these aspects are thoroughly treated.

Minor comments:

p 8759: "ground-based instruments cannot contribute to the study of transboundary pollution". I don't think this is fully true as measurements in clean air regions can provide valuable information on episodes of long-range transport of pollution

C2623

R: The sentence has been changed to : “However, they are unable to resolve and track pollution plumes in space and time and can only partly contribute, for instance, to the study of transboundary transport of pollution”

Section 3.1: How have the background measurements been selected?

R: The background measurements were selected to lie nearby, for their similar brightness temperature and ozone as the target spectrum. The additional signatures in the spectra could this way best be revealed in the difference. The sentence was changed to: “These were calculated by subtracting from the two target spectra within the plume a spectrum recorded nearby in background conditions (grey lines in Figure 1), which was chosen to have similar surface temperatures and ozone and humidity concentrations.”

P 8766: “likely indicating that the plume is so well confined at low altitude” – I don’t understand why the fact that the sensitivity of the measurements to the surface is enhanced can be used to deduce that the signal does not come from higher altitudes – why is that the case?

R: See above. The sentence has been changed to “If the emitted fire plume was in the boundary layer, this higher sensitivity would be a good demonstration of IASI probing the lowermost atmospheric layers.”

Section 3.2: in all respect: typo: respects $\mu$

R: This has been corrected

Section 4: found to be relatively high with expectations – I don’t understand this sentence

R: We meant here in comparison to existing literature. The sentence has been changed accordingly

Fig. 3 and Fig. 5: Why is the relative strength of the NH<sub>3</sub> lines different in measurements and simulations? Does this have an impact on the uncertainty of the retrieved

C2624

profiles and columns?

R: This is an effect of the radiative transfer in the region of the quasi-saturated ozone band. This has no impact on the results, as it is properly accounted for by fitting simultaneously the ozone profile. Furthermore, care was taken here for the ammonia retrievals to select channels which were outside the ozone band, where interferences are less.

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 8757, 2009.

C2625

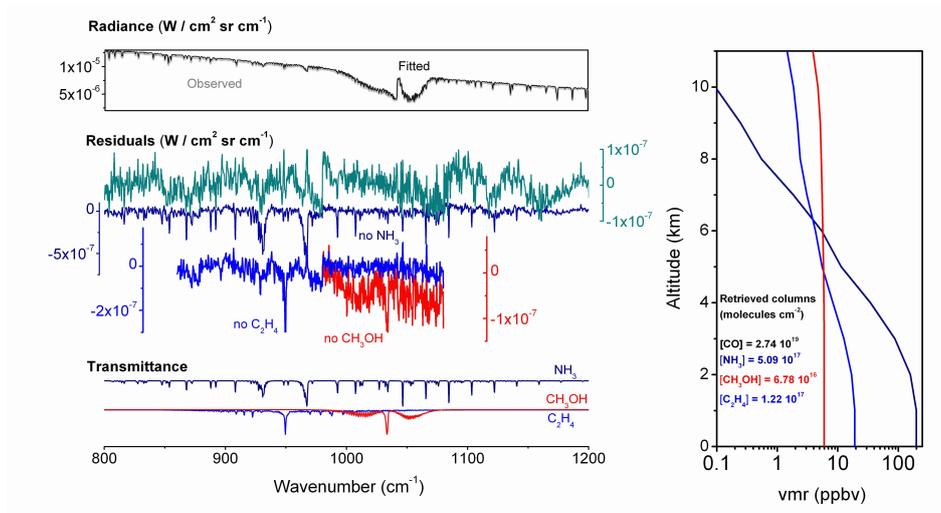


Fig. 1.

C2626

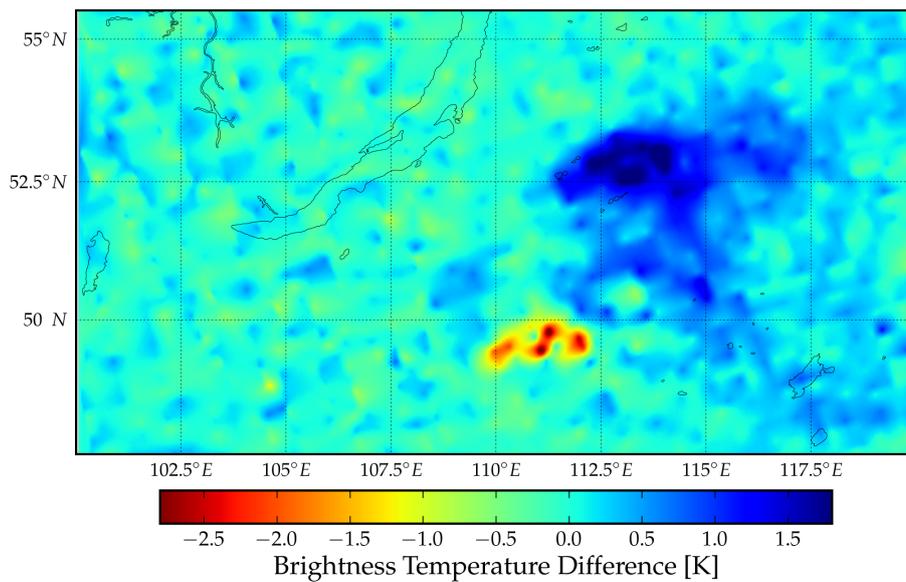


Fig. 2.

C2627