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Atmospheric Chemistry and Physics Discussions

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

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The authors report on the ability of the IASI instrument to probe various reactive species in biomass burning plumes. They selected two strong fire events to support their analyses: fires that occurred in Greece in August 2007 and fires that occurred in Siberia and East Mongolia in spring 2008. They show that spectral signatures of ammonia, ethene, methanol, formic acid, peroxyacetyl nitatre and acetic acid, attributed to fire emissions, can be observed and assigned in specific IASI spectra. They also show that the IASI observations allow probing the spatial and temporal extent of the plume. Finally the authors derived an estimate of the total mass emissions for NH3, C2H4 and CH3OH for the Greek fires as well as enhancement ratios relative to carbon monoxide in order to give insight in the chemical processes occurring during the transport.

We thank the referee for his detailed review and useful suggestions. We have addressed all general and specific comments. Our point-by-point responses to them are provided hereafter.

General comments

1) The main concern about this work is the lack of details and precisions about the quality of the products retrieved. The authors derive atmospheric columns of several species that would be very interesting and promising but they do not give any information about the characteristics of the retrieval (errors, averaging kernels, degrees of freedom, etc). This information, especially concerning the errors, is essential in order that the reader can judge of the ability to retrieve significant products and can judge of the relevance of the products for the discussion about the total mass emissions and the enhancement ratios derived. Indications about the a priori columns (and/or profiles) and typical background concentrations would also be welcome to give an idea of the enhancement for each emitted species. In Fig. 5, which is the vertical significance of the profiles (right panel) provided? I can believe that total columns can be retrieved but I'm sceptic for relevant profiles.

Reply to general comment 1

The paper provides the initial steps in measuring short-lived species with IASI, which we think is an unexpected and important success of the mission. The retrievals have not been performed focusing on vertical profiles, but more on getting first coherent estimates of concentrations in the plumes, which are needed to support the analyses as a function of space and time.

The characterizations that the referee asks (e.g. averaging kernels) assume that adequate prior information was used, especially with regard to the a priori covariance matrix. This has not been the case in this work: simple ad hoc variance-covariance matrices with very large variability and unrealistic correlations (7 km with an exponential decay) have been assumed in order to capture the large signals in the plumes,

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starting from background prior profiles. The latter were either from standard atmospheres (NH3) or were global annual averages from recent chemistry models (C2H4, CH3OH). The use of more appropriate prior information (e.g. more variability in the boundary layer and less higher up) would of course provide a different view of the vertical sensitivity. The averaging kernels from the retrievals performed here are thus not representative from the real instrument sensitivity.

Furthermore we would like to stress that a detailed characterization even in normal conditions (biomass burning is another source of complications) is difficult and lengthy because of the different aspects affecting the radiative transfer in the boundary layer, among which thermal contrast is probably the most important. A first extensive analysis has just been achieved for NH3 (Clarisse et al., in preparation) and we now make reference to the latter where needed in the manuscript.

Overall, detailed descriptions of the retrievals set-up and posterior analyses was thus intentionally left behind in the first version of the manuscript, for the reasons exposed above but also not to add to the technical character of the paper. We agree, however, with the referee and with the editor (who also raised these issues) that the elementary aspects of retrievals and of the errors were missing in the manuscript. The revised manuscript has been updated in that respect:

- A full paragraph giving details of the retrievals has been added (end of section 2.3.). It includes a description of the a priori profiles and an explanation of the ad hoc prior 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 NH3 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.). We come to the conclusion that the

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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, as rightly pointed out by the referee. 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 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) . . .."

2) The authors state that several species can be firmly detected from IASI spectra (p8768, line23). The spectral signatures are clearly visible in the residuals for NH3, C2H4, HCOOH, PAN in the examples chosen (Figs 3 and 5). However, the case of methanol is much less convincing. The Q-branch is very close to the noise level of the residuals and details about the retrieval would certainly help to determine if this species can be detected firmly or not. Are the two spectra chosen the best for this trace gas? Clarify the noise level?

Reply to general comment 2

We agree that the Figures did not bring sufficient visual evidence for the presence of CH3OH. We have changed them to better highlight the spectral residuals (which are now on a different scale for each species). The CH3OH Q-branch is now better appearing (especially in Figure 5 where the entire band P, Q and R band structure is seen: see attached figure for an example), although it remains close to the RMS of the fit. Regarding the latter, one should point out that it is about a factor of 2-3 larger

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than the real instrument noise, suggesting that the absorption strength of the different species is not negligible.

Figure 4 was also slightly modified by adding the spectral residuals with all species included (the 'best' spectral fit), in order to highlight the strength of the remaining absorption features.

As for the concern of the referee if the spectra are well chosen: yes, these are typical spectra for the fire events analyzed here. In more recent plumes, stronger signatures were detected (even by eye on the raw spectrum), of course giving us confidence on the results presented here. Furthermore, despite their weak contribution to the spectra, quasi-global distribution of CH3OH are in the process of being achieved using IASI.

As mentioned in the first point, the authors did not give enough information about the quality of their retrievals in order that we can evaluate the relevance of the discussion. If the two main points are addressed and if the results confirm the relevance of the measurements for the use that is done in this paper (total mass emissions and enhancement ratios), the work proposed would be of interest and would show the ability of IR nadir sounders to probe reactive species during strong pollution events. If these conditions are filled, the paper will be suitable for ACP publication.

R: We hope that with the additional information and the changes made to the manuscript (including the figures), we have brought convincing evidences on the capabilities of IASI to probe the short-lived species.

Specific comments

1) The authors should mention the atmospheric lifetime of the different measured species.

R: A range of lifetimes is provided (last paragraph of section 3.1.): "...are well known relatively short-lived biomass burning products (Andreae and Merlet, 2001), with lifetimes in the boundary layer ranging from a few hours (NH3) to several days (CH3OH),

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increasing for all species to several days/weeks at higher altitudes"

2) Part 2.2: How well is known the surface emissivity? Which is the impact of an error on this parameter in the method applied to detect plumes?

R: The surface emissivity is taken in this work from the MODIS database, as an average of the different TIR channels. It is thus a scalar value depending on the surfaces contained in each FOV. The method works fine in most cases, with slight deviations that are accounted for by fitting surface temperature (resulting in a very small error source). The main problems are found above desert and icy surfaces where a spectrally varying emissivity prevails. This does, however, not apply for the scenes analyzed here.

3) Part 2.3: The level 2 operational products of IASI (temperature, humidity, pressure) are used for the retrievals. If any validation of these products exists, the authors should refer to.

R: A sentence has been added (first paragraph of section 2.3) on the preliminary validation of the temperature and humidity level2 products, with the associated reference: "The preliminary validation of these level2 meteorological products reports on an accuracy close to the mission objectives: for temperature the error is 0.6 K in the free troposphere, increasing to 1.5-2 K at the surface and in the upper troposphere with a bias of $\pm$ 0.5 K, while for the relative humidity the error is 10 % with a bias within $\pm$10% (Pougatchev et al., 2009)"

4) In the conclusion, the authors write p8773, line 9: "This result is a comprehensible demonstration that thermal infrared sounders are capable of probing the atmospheric composition down to the boundary layer in favourable situations (e.g. positive thermal contrasts)". The authors should mention that this ability also depends on the combination between the shape of the profile and the sensitivity of the measurement. For the species studied here, most of the information comes from the lowest layers of the atmosphere and it is possible to conclude that one probe these layers because the concentrations of the species are larger in the boundary layer than in the other parts of

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the atmosphere to which the instrument is sensitive.

R: We thank the referee for this comment. The sentence in the conclusion has been revised, also taking the editor comments into account to: "The analyses have suggested, using available information on the local thermal structure of the atmosphere, and assuming that the emitted plumes were largely confined to the lowest part of the atmosphere, that IASI was able to probe the chemical composition deep in the troposphere. If confirmed by more detailed forward and inverse simulations, this result would open extremely promising perspectives for identifying local to global sources of short-lived species and for monitoring air quality."

Technical corrections

P8766, line 14: it should be "(right panel of Fig. 1)" and not "(left panel of Fig. 1)" Fig. 2. in the color scale, the minus sign is missing before 3.0.

R: The corrections have been made.

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

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Interactive Comment

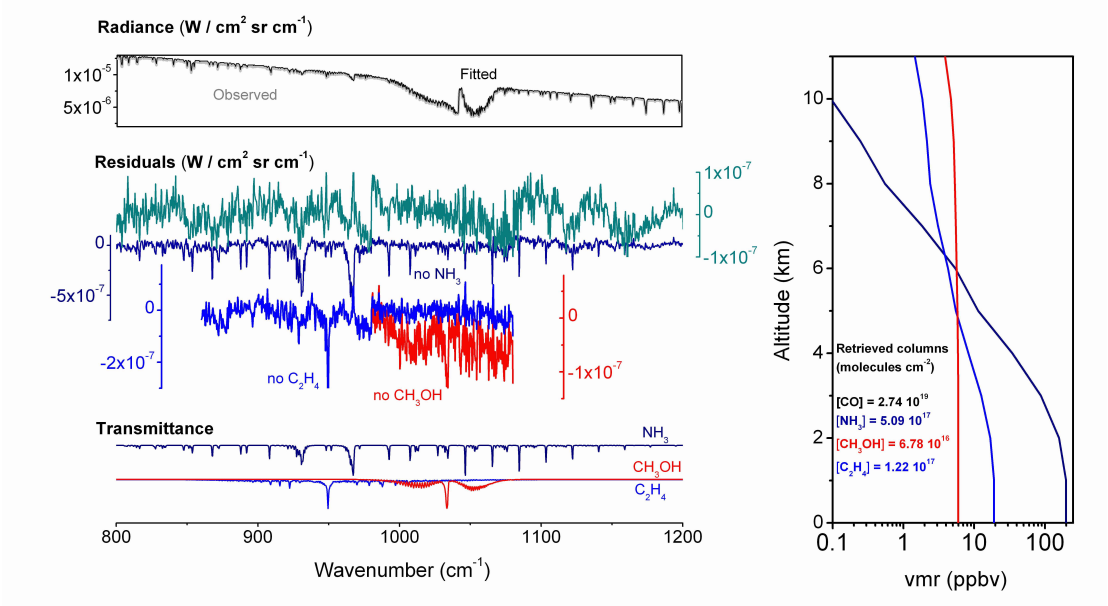

Fig. 1.

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