Atmos. Chem. Phys. Discuss., 12, C7479–C7487, 2012
www.atmos-chem-phys-discuss.net/12/C7479/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "FTIR time-series of biomass burning products (HCN, C_2H_6, C_2H_2, CH_3OH, and HCOOH) at Reunion Island (21 S, 55 E) and comparisons with model data" *by* C. Vigouroux et al.

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Received and published: 26 September 2012

Answers to Referee 2

The manuscript is well written and most of the methods used and analyses performed are clearly explained. The paper provides a good reference for the retrievals from FTIR at La Reunion, which have already been used in several studies. Therefore, I recommend publication with minor corrections and clarifications, following the specific comments listed below.

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Specific comments:

My main comment is that the data analysis regarding the seasonal and interannual variability remains somewhat too general. It could be improved by including a rapid analysis with respect to the emission inventories used for the model simulations for the same time periods (GFEDv2 for GEOS-Chem and GFEDv3 for IMAGES). These data are freely available and in an easily accessible format so that their analysis should not require significant work. It would help understanding the variability and the relative contributions from different ecosystem burning (available for GFED3 at least), an information that is critical for emission factors analysis. My second major comment is that although correlations to CO are shown, the comparison of the model simulations to that species are not shown or discussed. In my opinion, the analysis of the enhancement ratios would be more efficient if the reference (i.e. CO) was also evaluated with respect to observations.

We thank the referee for his/her useful remarks. As detailed further below, the revised manuscript now includes (a) a short analysis of forest and savanna emissions in both GFED2 and GFED3, and (b) a comparison of both IMAGES and GEOS-Chem results with FTIR measurements for CO. We have also taken into account most of the specific comments in the revised manuscript. We answer specifically to those comments below.

Detailed comments:

Section 2.2.1: p. 13741: The treatment of water vapor and the use of “a priori” is not clear to me in the part (l. 17-18) and the following of the section. Water isotopologues are 1st retrieved and then these retrievals are used in the retrieval of the other targeted species. But since the authors mention that the water retrievals are used as “a priori”, does it mean that they are fitted in parallel to the other species in the 2nd step of the retrieval?

Yes, the preliminary profile retrievals of the water isotopologues (and O_3, $CHClF_2$,

CCl_2F_2 for acid formic) are then fitted in the target species retrievals, but only with a scale factor (only the total columns of the interfering species can change, not their profiles).

So, in the first step, from a single annual a priori profile of water vapor, an individual vertical profile of water vapor per spectrum is retrieved. This profile is then used as the a priori in the next step (retrieval of target species). It could be "fixed", but usually an additional scaling of this a priori profile gives better spectral residuals because it compensates for small discrepancies between the spectroscopic parameters of water vapor in the first-step microwindow compared to the parameters in the microwindow selected for the retrieval of the target species. To clarify this, we changed the previous text "These individual retrieved profiles were then used as the a priori profiles to be scaled in the retrievals of the target species" with "These individual retrieved profiles were then used as the a priori profiles in the retrievals of the target species; they are again fitted, but now with only one scaling parameter".

Section 2.2.2, p. 13743: define WACCMv5, at least mention that it is a model. At the end of the section it would be interesting to discuss the possible biases due to the lack of sensitivity to the surface layer.

We have defined WACCMv5 in Section 2.2.1 (p. 13742 of ACPD version), and a web page was given. The lack of sensitivity to the surface layer is an issue mainly for HCN, and is mentioned p. 13759. As we do not know the "true" profile of HCN at Reunion Island, we do not give numbers to the possible biases. But, the information on how a "true" profile would be biased due to the FTIR sensitivity is contained in the total column averaging kernel of the target species that we give (Fig. 3), and this averaging kernel can be used by a reader that would have a profile to compare with our FTIR data. In our paper, we do apply our averaging kernels on the model profiles, and the biases that we obtain is visible on Fig. 4 (circles compared to solid lines).

Section 2.3, p. 13746: The authors introduce the Svar matrix but it is not clear

why this can not be used as a priori variability Sa (since authors mention that there is no valuable information on this in the previous section). Also, on what bases was the 4km correlation length chosen?

It occurs quite frequently that the Svar matrix is not used as the regularization matrix, and that either Tikhonov regularization is used, or some diagonal ”ad hoc” Sa matrix. This often happens because a good Svar matrix is not available, and /or to increase the stability of the retrievals (one easily obtains oscillations in the retrieved profiles if the natural variability leads to a constraint that is too weak). In our case, Svar is poorly known since we only have aircraft data, even not located at Reunion Island. Therefore, we have decided to use Tikhonov regularization. However, for the error budget, instead of giving no error at all for the smoothing (which is indeed advised by Rodgers (2000), in case no reliable Svar matrix is available), we have preferred to use the “poor” information we have to give at least an estimation of this error. The 4km correlation length is an “ad hoc” choice, based on what we found for the species HCHO using simulations from IMAGES (Vigouroux et al., 2009).

For clarity, we have added in the new manuscript that our smoothing error is only a “best estimate” based on our current knowledge, and that this error should be improved once a better climatology becomes available. We have added in Fig. 1 the variabilities associated with the a priori profiles as reprensented in Svar (as requested by the referee), but also the mean profiles and variabilities coming from our FTIR measurements. So, we can have a qualitative “a posteriori” evaluation of how good our estimation of the smoothing error is. Note that our text on the choice of regularization has been reduced to shorten Section 2 (cf. request of Referee 1).

P. 13747: For the interferences, the authors mention specifically the error associated to temperature, and also to the interfering species. What about water vapor? Can the specific error associated to this major interference be estimated precisely?

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Indeed, we give in Table 3 (Table 2 in the new manuscript) the total random error only, but it was calculated as the quadratic sum of all random errors, the error associated with each interfering species (including water vapor) being calculated individually, using Eq. 3 of the ACPD version (Eq. 3 has been removed in the new manuscript – see answer to Referee 1). We did not want to give the individual random errors to avoid entering into details. We found, for the main interfering species (H_2O for HCN and C_2H_6; HDO for HCOOH; $H_2^{18}O$ for C_2H_2; H_2O and O_3 for CH_3OH) an error of about 0.4-1%, except for C_2H_2 where the error due to $H_2^{18}O$ can reach 5-10%. Note that these relatively small errors are obtained with an S_b matrix constructed with a variability of 10%. If no preliminary retrievals of interfering species were made, this variability would have to be increased (especially water vapor is highly variable), and the resulting errors would be much larger. Of course, this 10% variability value is only an estimation. In Vigouroux et al.(2009), the S_b matrices have been derived from the estimation of the error budget of each interfering species that has been retrieved beforehand, but this paper concerned only HCHO, so only 3 interfering species. In the present paper dealing with 5 target species, we have decided that it is too much effort to do such a precise evaluation of S_b, especially when considering that the noise and the temperature errors always dominate the total error budget.

Section 3: Although the authors give a reminder of the results of another study for CO, it would be important for the understanding of the enhancement ratios to provide timeseries and comparisons to models for CO as well in Figure 4.

We have included the FTIR time-series of CO, together with the GEOS-Chem and IMAGES model runs for CO.

Note that the goal of the paper is not to compare the 2 models, so we do not discuss the differences that appear between both CO model datasets.

The discussion of the seasonal and interannual variability could be more specific if related to variability in the GFED (v2 and v3 used for the model simulations) in-

ventory in the main source regions for the considered time periods (with another figure). This would also help discussion in Section 4 (p. 13751 for instance). p. 13753: using GFED could also allow the identification of vegetation types for the analysis of enhancement ratios in this section (specific ecosystem could be more easily related to EF numbers in the literature). For C2H2, could a shorter lifetime explain the discrepancies (with different chemical evolution)?

We have added a comparison (Fig. 5) of forest vs. savanna emissions of CO according to GFED2 and GFED3 for the years of interest. See the new text added also in Sect. 3 and 4. Unfortunately, the use of GFED does not allow a straightforward identification of the vegetation type(s) impacting the FTIR measurements, because in the more recent GFED3 inventory, about as much emissions originate from forest fires as from savanna fires in Southern Africa. A more detailed study using backward trajectory in combination to GFED3 would be helpful, but this is beyond the scope of the present study. For C_2H_2, we have discrepancies with the references given in Table 4 (Table 3 in the new manuscript), but our results are in agreement with Ferek et al. (1998) and Paton Walsh et al. (2010), so it is hard to conclude. We have added a text in Sect. 4 to make it clearer to the reader that our correlation ratios are obtained using all FTIR measurements within the August-November period, without any distinction according to the origin of the air masses, i.e. according to the different types of vegetation.

Section 5: GEOS-Chem and IMAGES are two different models and different emission inventories are used for the simulations: a discussion of the possible implications is necessary.

Our aim was to compare our FTIR data to model calculations. It happened that IMAGES did not provide HCN, and for this reason, we included GEOS-Chem results (this was mentioned in the introduction). We don’t think it necessary to compare both models as the comparisons with the data are helpful and lead to the same clear conclusion: the GFED2 and GFED3 inventories seem to be underestimated in late September and October in the southeastern Africa-Madagascar region. In the ACPD version, we wrote

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in the conclusion section how GFED2 compares to GFED3 (GFED2 is actually lower than GFED3 in October). In the new manuscript, a comparison between GFED2 and GFED3 emissions is added, which supports this statement.

What meteorology is used to drive the IMAGES CTM?

Meteorological fields in IMAGESv2 are obtained from ECMWF ERA-Interim analyses. This information has been added in the new manuscript.

Why not use GEOS-Chem for the organic compounds as well?

As explained above, the purpose of the paper was not to compare the two models, but instead to compare our new FTIR data to some model results.

For both models, enhancements compared to CO are discussed but this assumes that CO in the models is correct? Are both models consistent in terms of CO?

A comparison of the model results with FTIR CO is included in the revised manuscript. Note that the analysis of modeled vs. observed enhancement ratios does not necessarily require that the modeled CO is correct. The fact that the discrepancy between model and data is more pronounced at the peak of the biomass burning season (October) for all species suggests that the main reason for the discrepancy is an underestimation of carbon emissions. This explains our conclusion (Sect 5.2.2, p. 13760 of ACPD version) that CO emissions are also underestimated in GFED3. We follow nevertheless the recommendation of the referee to include an additional plot to better clarify our point (CO time-series added in Fig. 4). We see in this additional plot that both models underestimate CO, especially in October. For HCN, we found that the enhancement ratio was too low (compared to our FTIR value, but also to the references given in Table 4). But if the smoothed model is compared to FTIR, even the correct enhancement ratio would not be sufficient to remove the discrepancy in October, confirming the underestimation in the emission inventory.

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Some discrepancies between model and observations depend on the season: a link to the origin of the air masses, their age, and the vegetation type burned may help understanding what can be learned about the emissions.

We agree. Backward trajectories would help to go further in what can be learned from our FTIR data. But at some point, one has to fix a limit to the scope of this paper. So, we hope that this could be investigated in the future.

For methanol comparisons: any idea why the IASI constrain is not enough to match the FTIR measurements? Do both datasets agree for this time period? Is it due to the inversion procedure (errors, etc.)?

Monthly means comparisons between IASI at Reunion Island and our FTIR data are shown in Fig. 11 of Stavrakou et al. (2011). Considering the very large error bars of IASI, the datasets were considered to be in agreement, but indeed, the amplitude of the seasonal cycle in IASI is lower than in the FTIR data, and the value for October is 15-20% lower in IASI. This suggests that IASI may underestimate CH_3OH at this period in the Southeastern Africa-Madagascar region. This text is added in the ACP version.

Technical comments:

p. 13745: avoid "..."

Not here anymore (Section 2 shortened).

Figures: Figure 1: it would be interesting to plot the variability of the a priori profiles.

Done. We have added in Fig.1 the variability used for the smoothing error calculation (square root of diagonal elements of Svar); but also, for comparison, the mean and standard deviation from the FTIR measurements themselves.

Figure 2: The corresponding DOFs could be provided in the title.

Done.

Figures 2 and 3: Why is there a sharp gradient in the C2H2 AKs around 20km? (This needs to be explained in the text).

We suspect that it is due to numerical issues when the volume mixing ratio (vmr) becomes too small (10-13 – 10-14 ppv), because we observe the same behavior for HCOOH around 32 km, when the vmr of HCOOH also decreases to these small values. We prefer not to include such a remark in the text because: 1) referee 1 asked to shorten this section; 2) 99.8% of C_2H_2 is located below 17 km, so this sharp gradient has no impact on the total columns presented and discussed in the paper.

Figure 4: should add CO in the list of species – for reference.

Done.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 13733, 2012.