

Interactive comment on “ACE-FTS observation of a young biomass burning plume: first reported measurements of C₂H₄, C₃H₆O, H₂CO and PAN by infrared occultation from space” by P.-F. Coheur et al.

P.-F. Coheur et al.

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We thank the referee for his review of the paper and very useful comments and suggestions, which have helped to improve the paper. All the comments have been addressed and information has been added where requested. Below is or point-by-point responses to the referee's comments.

General Comments

The paper describes the analysis of a particular occultation sequence from the ACEFTS instrument where the absorption features of a number of biomass-burning

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related molecules have been detected in the spectra. The authors are careful to state which molecules they believe they can detect and which are doubtful, and the spectral signatures of the former look convincing. Comparisons are made with earlier in situ measurements within such plumes but, given the unusually high altitude of this particular plume, it not possible to draw any firm conclusions as to whether or not the retrieved composition is in line with previous observations.

Data from a chemistry-transport model are used in support of this study but only in the limited sense of identifying the source of the plume as local. This, I have to say, is not convincing: the measurement location (Fig.3b) seems outside the predicted local biomass burning in Africa and, judging by the shape of the plume, the wind seems to be blowing the CO in the wrong direction. On the other hand, given that this seems such a complete chemical model, why not include the modelled CO and other chemicals for the plume and "background" profile locations in the tables and figures for comparison?

Reply

We thank the referee for this useful comment. We had a closer look at the model data and in particular to the wind fields for the three days period considered. First of all, we would like to mention that the region was indeed subject to intense biomass burning during the period in October considered, as confirmed by the ATSR fire counts, with spots located very close to the occultation. However, the referee is right when he says that the principal circulation patterns blow CO from these fires mostly the other way round (e.g. to the West). Despite of this, the model calculations still find that the dominant contribution to the total CO in the upper troposphere above 300 hPa (~10 km) originates from this fire region, probably as a result of vertical uplift or after the plume has circled around and being progressively lofted before reaching the occultation point. To make this clearer, we have replaced Figure 4, which showed global model distributions, with a new figure highlighting at the measurement location, the agreement between the CO observed and model profile, and the relative contribution to the latter. Section 3.1. was modified accordingly.

As for the comparison with modeled and observed vmrs, for CO and other species, we agree with the referee that it would indeed be interesting information. The comparison is now shown in the revised Figure 4 for CO of the target occultation (left panel). As expected, what this figure reveals is that we are dealing here with a rather remarkable event in the upper troposphere, which is not reproduced accurately by the model (the model misses the enhancements at 12 km). We therefore believe that more detailed and quantitative comparisons would require more sophisticated photochemistry models within biomass burning plumes, such as those referenced in the paper. This is, however, beyond the scope of this paper, whose primary objective is on the measurement of previously undetected species in the ACE-FTS spectra.

Comment

There have been previous papers on detection of biomass burning signatures using the ACE-FTS instrument, by different groups, but since these are probably the first reported detections for some molecules from satellites, a new paper is justified. However, the major deficiency in this paper is the lack of any sort of error analysis of the results given. Even with a simple least-squares fitting procedure it should be possible to assign some statistical error bars to the tabulated results and figures.

Reply

We agree with the referee that error estimations are missing. Error bars are now shown for the CO retrievals in Figure 4, but were not added for the other species in Figure 6, for the sake of clarity. For the latter, we have adopted the referee's suggestion of adding the statistical error from the retrievals in Table 2.

Specific Comments

Novelty: There are a number of differing claims to the novelty of the data, from "first reported detections using infrared occultation spectroscopy from satellites" (Abstract, line 16); "first reported measurements from infrared occultation sounders" (p7916, line

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4); and "first reported simultaneous measurements by means of infrared spectroscopy from satellites" (p7919, line 3). All of which have subtly different meanings (eg as far as I know, there has been only one satellite infrared occultation instrument - ACE; unless you also include ATMOS which, being a shuttle experiment, was not strictly a satellite instrument).

Reply

There have been recent measurements of NH₃, acetone and PAN from the MIPAS satellite instrument or from the MIPAS-balloon instrument, which have been adequately referenced in the paper. Both operate in emission but also the infrared spectral range. H₂CO is routinely observed from nadir-viewing satellite instruments operating in the UV spectral range. For C₂H₄ these are, as far as we know, the very first measurements from satellite instruments. For these reasons, to be objective on the novelty of our results, while including in one sentence all species investigated, we found it necessary to keep the terms "infrared"; "occultation" and "satellite".

Comment

Vertical Resolution: On p7910 line 13 the average vertical resolution of ACE-FTS is given as 4km, yet in Fig.6 the profiles are plotted at 1km resolution and on p7917 it is claimed that the maxima for various species can be located to within the same 11-12km interval. Please clarify.

Reply

The vertical resolution of ACE-FTS depends on the vertical sampling, which varies from 2 km in the troposphere to 4 km in the mesosphere, but also from one occultation to the other. The version 2.2. data are also provided on a constant 1 km vertical grid. This is achieved using an a posteriori piecewise quadratic interpolation scheme method. This is clarified in the revised manuscript (section 2.1., second paragraph).

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HNO₃ retrieval: p7911, line 4: "it is worth pointing out that we do not use [the standard ACE-FTS L2 HNO₃ product]". Why not? Is there something wrong with it?

Reply

Indeed in our analyses of HNO₃ distributions, we found that version 2.2. showed spurious vmrs in the troposphere. The reason was investigated and finally an independent set of microwindows, all located within the nu5-2nu9 absorption bands have been selected to provide improved results for the lower altitudes.

Comment

Ozone: Why is ozone not retrieved? Ozone enhancement is considered a good indicator of photochemical processing in biomass plumes so a low (background) ozone concentration in the plume would have been more evidence that this is indeed a recent plume.

Reply

Ozone is retrieved operationally from the ACE data. For the particular occultation discussed in the paper, we do indeed not find evidence of strong enhancements: at 11.5 km, the ozone vmr is 69 ppb for the background occultation and 95 ppb for the target occultation (enhancement of 37 %) but the shape of the profile is in no way similar to the other species. This could be a further evidence for the recent character of the plume and we thank the referee for pointing this out. The argument has been added at the end of the third paragraph in section 3.4.

Comment

Apodisation: Are apodised or unapodised spectra used? The ACE-FTS team generally prefer unapodised but, particularly when looking for small signals, the "ringing" effect caused by the truncation of the fourier transform can produce spurious signals if not modelled correctly - I do not say that that is what is seen here but it should be stated whether or not apodisation is applied.

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Reply

Unapodised spectra have been used. This has been clarified (second paragraph, section 2.3.). We understand the referee's comments with regard to modeling the ILS but in the present case an accurate formulation was used (accounting for finite FOV, additional sources of self-apodization), as prescribed by Boone et al. (2005). It produces, for well isolated lines, residuals below the noise levels. Accordingly, we do not believe that the modeling of the ILS can account for the observed residuals.

Comment

Interfering Species: On p7913 line 9 it is stated that the v2.2 profiles of all interfering species are adjusted to provide the least best achievable fits. Unless some sort of constraint is applied (particularly for the weaker absorbers in a particular spectral region) this is a dangerous procedure which may create more spurious features.

Reply

The procedure that we have adopted basically consists in using the version 2.2. profiles for all species, which were performed on a selection of dedicated microwindows. The profiles were readjusted in the spectral windows for the target new species only to improve on the quality of the fits and to isolate the spectral features of interest. It is worth pointing out that the profiles of the interfering species were only marginally different. We agree with the referee that if it wasn't the case, it could lead to some misidentifications. For the situation presented here, we do not think that there are ambiguities on the assignments, except maybe for NH₃ and C₃H₄. For these two species, we have, however, been relatively cautious in the identification.

Comment

Background Continuum: Is a background continuum also fitted for each spectral region? For the broader featured absorbers, such as PAN and acetone, this can make a significant difference to the answer.

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The molecular continua have not been adjusted. Their signatures extend over wavenumber ranges much larger than those considered here (even for PAN), and they can accordingly not be well isolated. In order to reduce the impact of such continua on the retrievals, both a baseline and a slope coefficient have, however, been adjusted in the spectral windows of interest.

Comment

Plume Origin: If the plume is assumed to be recent, ie of local origin, and is of sufficient strength to be lofted to high altitudes, it seems strange that its origin cannot be more convincingly located with the use of the ATSR data. (I assume these are actual fire detection data and not some climatological distribution).

Reply

ATSR fire counts (not shown in the paper) suggest that there are actually a fairly large number of fires in the area. We do not find it straightforward to assign a single event to the observed plume.

Comment

CH₃OH & HCOOH: On p7914 line 20 it is stated that the retrieved concentrations of these molecules "are consistent with the average mixing ratios earlier reported for aged plumes". However, in aged plumes I would expect the mixing ratios to be considerably reduced (due to mixing with background air) compared to a recent plume. Does this actually mean "consistent" in the sense that one might eventually evolve into the other?

Reply

The CH₃OH volume mixing ratios reported in Dufour et al. (2006) range from a few hundreds of pptv to 5 ppbv whereas the HCOOH reported in Rinsland et al. (2006) are estimated, based on a small number of profiles to be 0.94 +- 0.65 ppbv. At 12 km for the

present occultation, CH₃OH and HCOOH vmrs are 2.03 and 0.49 ppbv respectively, which are within the ranges of values reported in the two above-mentioned papers. We agree with the referee that mixing with background air would tend to reduce the vmr in aged biomass burning plumes but other factors are also involved (quantity of products emitted, photochemistry), which makes it difficult to draw firm conclusions on such a limited number of occultations.

Comment

Species not detected: p7916, lines 6-14: it would have been useful to have the detection limits for these additional molecules so that the absence of any detectable signal can also provide information on the upper values of the concentrations.

Reply

Giving a detection limit implies finding the most suitable microwindows, estimating the shape of the profiles etc; In the absence of previous observations from a satellite in this observing mode, such assumptions could lead to rather imprecise estimations. We therefore do not find it relevant to add such information at that stage.

Comment

Ethene lifetime: p7916 line 18: Mauzerall et al are quoted as giving a lifetime of half a day. In Table 1 of their paper, that value is given for 0-4km altitude. For this particular plume, wouldn't the 8-12km altitude figure of 1.2 days be more appropriate?

Reply

We agree with the referee. The sentence has been revised to "which as a mean lifetime ranging from half a day in the lower troposphere to 1.2 days in the upper troposphere"

Comment

Yokelson et al results: On p7918 it states that the Yokelson results are "measured just above a fire in Mozambique" which is a slightly misleading statement since it suggests

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a comparable situation to the ACE-FTS observations which could also be described in the same way. The Yokelson results (from their Table 2) are the "initial emission ratios" extrapolated back in time to the emission source, so not directly comparable with these (or any other) subsequent plume observations. Unfortunately Yokelson et al do not provide any information on the time evolution of these ratios so it is not possible to make any adjustment to their results, but it should be made clearer that these data differ in that respect.

Reply

This has been made clearer in the revised manuscript (page 7918). Furthermore, to comply with the referee's suggestion made below, we do not make reference to the Mozambique fire but more generally to the "study average value" of the Yokelson paper.

Comment

Sunrise or Sunset?: I understand that all ACE-FTS occultations are either at local sunrise or sunset but I didn't notice it mentioned which applies here - presumably has a significant bearing on the photochemical processing in a recent plume. I assume that the "ss" notation for the occultations studied here refers to sunset, but I am not familiar enough with the orbital mechanics to know whether a "sunset" from the instrument point of view necessarily corresponds to a local sunset.

Reply

The ss notation indeed refers to sunset. This has been clarified in the text (page 7911) and in the caption of Figure 1 and Table 2.

Comment

Applicability to other cases: The plume is clearly untypical in reaching such high altitudes but, having read the paper, I am still unclear as to whether this is just coincidental or whether such a clear signal is *only* detectable because it is at high altitude? Clearly the absence of cloud is a prerequisite, but would line-broadening, water-vapour, haze

production etc prevent such a retrieval at low altitude? I accept that the authors have just analysed a single case, but I think it would be helpful to make some further statement as to the potential of ACE-FTS as an instrument for routinely measuring such biomass burning products.

Reply

At the stage of writing, it was not obvious to know whether the analyses could be applicable to many other cases or not. We have recently tested the possibility to obtain more global distribution for C₂H₄ and H₂CO. Preliminary results suggest that retrievals can be successfully performed for a large number of cases. The resulting distributions in the upper troposphere bring interesting results, which will be discussed in a forthcoming paper.

Comment

Table 2: Apart from adding some 1 sigma uncertainties to the retrieved values, it would be useful to have the Mauzerall values here as well and, perhaps those from the model. Also, rather than take the single Beira case from Yokelson et al, I suggest using their "Study Average" values, plus 1 sigma variability (which usually incorporates the Beira values).

Reply

As explained above, the model reproduces the large scale features of the transport and chemistry well but misses more fine features associated with unusual events. As shown in Figure 4, for the particular occultation studied here, the comparison between measured and simulated vmrs within the plume lead to poor agreement. The simulated vmrs have therefore not been added in Table 2. On the contrary, we have followed the referee's suggestion to replace the Mozambique fire values of the Yokelson et al. paper by their "study average value" and to add the values for fresh, recent and aged plumes of the Mauzerall et al. study. The latter comparison for C₂H₂, C₂H₄ and C₂H₆ gives

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indeed further evidence for the recent character of the plume investigated here. The text in section 3.4. (last paragraph) has been slightly revised in order to account for these new values in the table.

Comment

Figure 3: Perhaps it is a problem with the colour scale, but it seems that the total CO over S America (a) is significantly larger than the biomass burning contribution (b), in which case: where does the extra CO come from?

Reply

CO is the result of incomplete combustion and is also produced by the oxidation of hydrocarbons; it has a lifetime of several weeks and is effectively transported within one hemisphere. In the upper troposphere, the CO background vmr is around 50-100 ppbv depending on location. The biomass burning contribution in S. America amounts 160 ppbv, which adds to the background values to give the total value of 250 ppbv in Figure 1.

Comment

Figure 5: It would be useful to have some quantitative measure of the improvement in the fit generated by the additional molecules, eg write the chi squared statistics before and after the fit on the figures.

Reply

The green residuals have been obtained by fitting the spectrum with the target species accounted for. In order to obtain the purple curve, only a forward calculation was performed, by keeping the results of the adjustment for all parameters, except for the vmr of the target species, which was zeroed. The chi squared can therefore not be compared this way. Nevertheless, the level of improvement is fairly well to be seen from the figures.

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