**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Ground-based FTIR and MAX-DOAS observations of formaldehyde at Réunion Island and comparisons with satellite and model data" *by* C. Vigouroux et al.

C. Vigouroux et al.

corinne.vigouroux@aeronomie.be

Received and published: 30 November 2009

We thank the referee for his positive comments that helped us to improve the manuscript.

General comments:

I recommend to re-structure the manuscript. Both FTIR and MAX-DOAS retrievals are based on the optimal estimation method, and information on this technique (basic retrieval strategy, averaging kernels, error budget) is spread over sections 2 and 3. I suggest to first introduce the OEM in general in a separate section, and to discuss the specific application of OEM to FTIR and MAX-DOAS in the following two sections.

Also, the construction of the a priori, commonly used both by FTIR and MAX-DOAS, could be moved to this general section. This would avoid unnecessary repetitions.

Reply: We have followed the referees' suggestion and a new section has been added (Sect. 2) which introduces the OEM, and gives common information such as a priori profile and S_{var}.

The same applies to the methodology used in sections 6.1 to 6.4... Here, I also recommend an introductory section explaining the statistical methods, the generation of "smoothed" profiles and vertical columns and the respective covariance matrices, and to describe how these methods are applied to the individual datasets in the following sections.

Reply: Here, we prefer not to use a common section explaining the generation of smoothed profiles and covariance matrices, because we did not use the same equations for Sects. 6.1 and 6.2. But, we have added a short introduction before Sect. 6.1 (now Sect. 7.1) which clarifies what is common to the following sections.

Specific comments:

Section 2.2: The retrieval strategy starts with the discussion of the averaging kernels, but information on how the state vector is calculated from the measurement vector y is missing. This is only mentioned later in the DOAS retrieval section (3.2). Please specify which quantities are represented by the measurement and state vector. Which vertical grid is used ?

Reply: These informations have been added (Eq. 2, and Sect. 3.2.1)

Section 2.2.2 It is not obvious why "information is not available" on the natural variability and thus on the a priori covariance of HCHO at the measurements site (P 15898, L. 26). The a priori has been constructed from the PEM-Tropics-B database, and this data could also be used to construct Sa. Later on (Eq. 3), the covariance constructed from this dataset goes into the calculation of the smoothing error. Aren't there other

reasons for the choice of regularization instead of maximum a posteriori, such as the advantage of having less constraints on the absolute value of the a priori profile?

Reply: We have to make a distinction indeed between the choice of the regularization in the retrieval and the a priori covariance matrix needed in the calculation of the smoothing error. As to the retrieval: In the FTIR case, an advantage was cited in the manuscript (P 15899, L4-6): "This regularization avoids the appearance of spurious oscillations in the retrieved formaldehyde profiles that occurred in the usual OEM implementation". This is indeed thanks to the fact that Tikhonov regularization constrains the solution to the shape of the a priori profile rather than to its absolute values. In the MAX-DOAS case also, Sa is not the natural variability of HCHO (P15905, L7-10): "The S_ϵ matrix being fixed, the a priori covariance matrix Sa can act as a tuning parameter (Schofield et al., 2004) and diagonal elements corresponding to an error of 70% have been empirically determined in order to ensure a good fit of the measurements". We have clarified this in the new section describing the OEM (Sect. 2) since it is very common to use Sa as a tuning parameter instead of the natural variability of the molecule, in order to have stable retrievals. For the calculation of the smoothing error (via S_{var}), we must use our best estimate of the natural variability at Réunion Island (PEM-Tropics-B). However, this information is not perfect since it concerns mainly marine measurements while Réunion Island is found to be affected by long-range transport of biomass burning products. This "imperfection" in our calculation of the smoothing error is mentioned in Sect. 6.3; P15917 L15-20 (now in Sect. 7.3). This was the reason why we had written that we didn't have information of the variability at Réunion Island (P 15898, L. 26). However, we have removed this sentence since it is indeed not the main reason for the use of Tikhonov regularization in our case.

Section 2.3: The measurement error S_ϵ is determined a posteriori by comparing the observed and calculated spectrum (P. 15902, L. 6). S_ϵ should only contain random errors, and the question arises if S_ϵ is influenced by any remaining spectral structures which might cause systematic errors (interfering species, instrumental line shape,...)

Reply: The referee is right in saying that some remaining spectral structures might influence the calculation of S_ϵ. However, in general and in the particular case of HCHO, these structures are of the same order of magnitude as the random noise, and can be positive or negative within the micro-windows. Thus, they may cause only a slight overestimation of the random noise and thus of the measurement error.

Section 3.1: A more detailed description of the DOAS analysis is necessary. In order to convince the reader of the quality of MAX-DOAS measurements, please provide more information on the DOAS retrieval procedures, i.e. typical errors of the HCHO dSCDs, typical residual RMS, detection limits, and also a figure with a sample HCHO retrieval.

Reply: Typical errors on the HCHO dSCDs appear in Fig. 6 page 15947 where examples of retrieval fit results are presented. In the revised version of the mansucript, we have included information on typical residual RMS (5×10^{-4}), detection limit (7×10^{15} molec/cm2 in slant column) as well as a plot with an example of a DOAS fit for HCHO.

Section 3.2: Usually, the measurement vector contains DSCDs relative to the zenith reference from the same measurement sequence. Here it appears that the DSCDs are relative to a measurement at noon, but there is a different amount of HCHO at noon than at the time of the measurement. How has this been resolved?

Reply: Actually our measurement vector contains DSCDs relative to the zenith reference from the same scan but it is a bit misleading in the paper. In Section 3.1 (now Sect. 4.1) 'Instrumental set-up and DOAS analysis', it was written that we used a daily noon zenith reference. It is true but in the profiling step, the DSCDs are re-evaluated using for each scan the zenith reference from the same scan. This second step is not mentioned in the paper. To avoid any confusion, the sentence 'We use a daily noon zenith reference' on line 2, page 15904 is removed in the revised version of the manuscript, and it is mentioned now in Sect. 4.2.1 that in the measurement vectors, the DSCDs of a given scan are relative to the zenith reference from the same scan.

It would be useful to mention that, in the DOAS community, the weighting functions are

commonly referred to as box airmass factors, Kij = dSi/dvj with vj being the partial VCD at altitude zj

Reply: Such a sentence is added in the revised version of the manuscript.

Apparently, the estimation of the aerosol profiles has been done 'by hand', by comparing measured O4 with modelled O4, generated using a set of pre-defined aerosol profiles. This approach is probably sufficient for an accurate simulation of the radiative transfer for the purpose of trace gas profile retrieval. However, to my knowledge, BIRA has developed a more sophisticated, automated aerosol retrieval model based on optimal estimation. Rather a question of interest: what is the reason for not using this aerosol retrieval algorithm?

Reply: The reason is very simple: this study on HCHO has been performed before the development in our Institute of a sophisticated and automated tool for the retrieval of aerosol properties from MAX-DOAS O4 observations.

Section 3.3.2: The authors argue that the small number of elevation angles (5) is the reason for a relatively low information content, and that measuring at more viewing directions could lead to an improvement. The fact that measurements at different elevation angles are not completely independent represents a principal limitation of the information content of MAX-DOAS measurements. Therefore, from my experience, an increase in the number of viewing directions does not improve the information content significantly.

Reply: We agree with Referee #1. In our sensitivity test where the noise on HCHO DSCDs is reduced and the number of viewing angles is increased (see lines 20-25, page 15907),we found that the main contribution ($\simeq$ 85%) of the DOFS increase comes from the noise reduction on HCHO DSCDs. The increase of the number of viewing angles has therefore only a limited impact on the DOFS ($\simeq$ 15%), as suggested by Referee #1. We added the following sentence: 'However, most of this DOFS increase is due to the noise reduction on HCHO DSCDs and not so much to the increase of the

C7626

number of viewing angles mainly because measurements at different elevation angles are not completely independent.'.

Technical corrections:

P. 15907, L. 14: "The information content is characterized through the calculation of the averaging kernel matrix" - replace "through the calculation of" with "by the trace of".

Reply: This sentence is removed in the new manuscript.

P. 15908, L. 11: Move "are presented in Table 4" to the end of the sentence.

Reply: Done.

P. 15917, last line: "interpolated at the measurement site" – replace "at" with "for".

Reply: Done.

Figure 15: The maps showing the CO tracer source strength is hard to read. Can you generate scaled-up maps with a zoom on Madagascar?

Reply: We have changed Figure 15, with a small zoom (-10 to-30° latitude; 20 to 60° longitude). We prefer not to zoom only on Madagascar because we want to illustrate that within 1 day, transport to Réunion Island is not coming from Africa.

───────────────────────