

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.

Anonymous Referee #1

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General comments:

The manuscript entitled 'Ground-based FTIR and MAX-DOAS observations of formaldehyde at Réunion Island and comparisons with satellite and model data' by Vigouroux et al. presents comparisons of HCHO vertical column densities derived by FTIR and MAX-DOAS measurements.

The paper is of excellent scientific quality. Apart from a direct comparison between FTIR and DOAS (which, to my knowledge, has not been published before), the retrieved formaldehyde amounts have also been compared to satellite measurements and results from a chemistry model. Using a particle dispersion model (Flexpart), the

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source of air masses enriched in HCHO and CO, as well as possible uncertainties in the chemistry and/or transport models could be identified. The intercomparison between the numerous data sets has been done with great caution, taking into account the differences in vertical resolution and sensitivity for different altitudes. These inter-comparisons have been performed on the basis of well established statistical methods as described by Rodgers and Connor.

MAX-DOAS is a relatively new measurement technique, and publications on the comparison of vertical profiles or tropospheric VCDs retrieved from DOAS trace gas slant column density measurements with independent instrumentation are yet sparse. Therefore the validation of MAX-DOAS data products is of scientific importance, in particular for the DOAS community.

To enhance the conciseness of the paper, 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 optimal estimation method in general in a separate section (including the respective equations currently mentioned in sections 2 and 3), 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 DOAS, could be moved to this general section (currently in section 2.2.2). This would avoid unnecessary repetitions.

The same applies to the methodology used in sections 6.1 to 6.4. In each of these, two datasets are compared using the same methodology, i.e. applying the respective averaging kernels to the datasets according to Rodgers and Connor. 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

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sections.

Specific comments:

Section 2.2:

The retrieval strategy starts with the discussion of the averaging kernels, but information on how the state vector \hat{x} 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 (i.e., vertical profiles and radiance spectra, respectively). Which vertical grid is used?

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 S_a . Later on (equation 3), the covariance constructed from this dataset goes into the calculation of the smoothing error. Aren't there other reasons for the choice of regularisation instead of maximum a posteriori, such as the advantage of having less constraints on the absolute value of the a priori profile?

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...).

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

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residual RMS, detection limits, and also a figure with a sample HCHO retrieval.

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?

It would be useful to mention that, in the DOAS community, the weighting functions are commonly referred to as box airmass factors, $\mathbf{K}_{ij} = dS_i/dv_j$ with v_j being the partial VCD at altitude z_j

Apparently, the estimation of the aerosol profiles has been done 'by hand', by comparing measured O_4 with modelled O_4 , 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?

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.

Section 4: The sentence "The HCHO columns have been calculated with and without accounting for the averaging kernels of the measured FTIR and MAX-DOAS columns." is out of context, the methodology for accounting for different vertical sensitivity is only

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explained later in the manuscript.

Sections 6 and 7:

Apart from the recommendation to re-organise section 4 as outlined in the general comments, I have no specific comments. The results are presented in a very comprehensive and conclusive manner, under consideration of the differences in vertical sensitivity of the different datasets.

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".

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

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

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?

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 15891, 2009.

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