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Atmospheric Measurement Techniques
Discussions

Interactive comment on "NDACC harmonized formaldehyde time-series from 21 FTIR stations covering a wide range of column abundances" *by* Corinne Vigouroux et al.

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This paper describes the production of a harmonised data set for HCHO column abundances from 21 FTIR stations located across the globe. First of all, I must commend the authors for pulling this off. I cannot imagine it was an easy task. Bringing together the HCHO measurements from these different stations/groups is an important achievement, and it will be a valuable resource for modelling studies and for satellite validation purposes. It is a great step forward. I urge the authors to create an online repository where the data can be download easily by others. Overall I recommend the paper for publication, the science and methods are sound, the results important, and it is well-written. I only have minor comments that need clarifying.

We warmly thank the referee for his/her kind supporting words. Concerning an online repository where the data can be downloaded, this is currently under discussion within the InfraRed Working Group (IRWG) community, and a final decision will be taken at the next IRWG meeting in June. The data will be very likely downloaded in the public NDACC repository. In the meantime, the whole data set is provided on request by myself (corinne.vigouroux@aeronomie.be), or station by station by the individual PIs. I take this opportunity to remind that if any FTIR data is used in a publication, even when the data is released in the public NDACC database, the appropriate PI has to be contacted. If the use of FTIR data is a significant contribution to the publication, a co-authorship should be offered. Otherwise, in agreement with the PI, acknowledgements can be sufficient.

Minor comments:

Please ensure all figures are large in the final version of the manuscript – they are very small and difficult to resolve; it is frustrating. Maybe they could be enlarged by breaking them into separate figures.

The figures have been enlarged in the AMT version.

I noticed that in Table 1 the observing period is very long for some of these stations (e.g., Ny-Alesund). Could the authors possibly comment on the instrument stability and performance over such long periods, and if it affects the HCHO retrievals at all? HCHO is difficult to retrieve is not?

Usually the instrument stability is checked by making regular cell measurements, which allows to verify the good alignment of the instrument (Hase et al. 2009; see also Sect. 2.1 in the present AMTD version). The instruments are re-aligned when a misalignment is detected. Other type of degradation, e.g. of the mirrors, can easily be seen in the decrease in signal to noise ratio (SNR) of the spectra. This decrease in SNR has a direct impact on the precision of HCHO columns (dominant random error source). Therefore, the precision can indeed vary during the time-series period, but the information is anyway provided in the data sets which include random uncertainties associated to individual measurements. The mirrors need to be regularly replaced to avoid a too low SNR. The data sets are also quality controlled after the retrieval process. A too low SNR would lead to a bad root-mean-square (RMS) of the fit, and a threshold on this RMS is used at each station to reject the bad quality spectra. The spectra that do not pass the quality assurance (instrument alignment, RMS,. . .) are removed from the data sets, leading to small gaps in the time-series as seen in Fig. 5.

Page 7, line 25. The a priori HCHO profile. The approach used here seems sensible, but how sensitive are the retrieval total columns to the a priori – especially as the DOFs is low.

The effect of the a priori profile (and Svar matrix), is calculated in the smoothing error, which has a random but also a systematic component. The systematic smoothing error component, more closely related to the a priori profile itself, was found to be non-significant in our study (1-2% in most cases, therefore dominated by the other systematic sources, which range from 12 to 26%), as was (too shortly, indeed) written p.11, l.10-11.
This small systematic uncertainty was obtained using the following equation:
(I-A) (xa - <x>) (xa - <x>)^T (I-A)^T,
which accounts for the bias of xa, i.e. which accounts for the fact that xa might be different that the expected real <x>, following von Clarmann (2014).
The xa - <x> is obviously not known (otherwise, <x> would be chosen as the correct a priori in the retrievals). Therefore, we had chosen in our AMTD version to use the diagonal elements of the Svar used in Eq. 4 (for the random smoothing error component), as an estimation of xa - <x>.
However, Referee#3 also asked for more discussion about this systematic uncertainty.

We have therefore added the equation above to the new manuscript. We have also decided to use larger values than the ones from Svar for the revised manuscript: we considered the systematic smoothing error that would occur if the a priori profile is differing from the real <x> by -50%, -20%, -10%, +10%, +8%, +5% for the ground-4km; 4-8km; 8-13km; 13-25km;25-40km; 40-120km layers. The values have to vary with altitude to induce a different a priori profile shape: if 50% is used at all altitudes, the a priori profile is then different from <x> by a simple scaling factor, and the systematic smoothing error is close to zero. Using the above values, we obtain systematic smoothing errors from 1 to 9% (median value of 3.4%), which is still small compared to other systematic error sources.
These values rely on the fact that the model WACCM a priori profile shapes are not too far from the reality, which should be the case: due to the known short lifetime of HCHO and its production at or near the surface, we expect that the mean profile peaks at the ground.

This is, as for the random smoothing part, only an estimate of the smoothing systematic error. As discussed in von Clarmann (2014), one would prefer even to not give these smoothing errors at all. We prefer to give them in our paper to provide to the reader as least an idea of the impact of the smoothing in the precision and accuracy of our FTIR HCHO measurements. But these smoothing errors are not provided in the .hdf files that are delivered to the public. When making model or instrument comparisons, the appropriate use of the averaging kernel and a priori profile information (provided in the .hdf files), following Rodgers et al. (2003), allows the user to take implicitly into account the smoothing uncertainty. This means that, for satellite or model comparison, if Rodgers et al. (2003) is used, there cannot be some different systematic biases at different stations due to different xa-<x>.

T von Clarmann, Smoothing error pitfalls, Atmos. Meas. Tech, 7, 3023-3034, 2014.

Figure 3. The use of atm16 is clearly necessary; HITRAN 2012 needs some corrections...

Indeed. Spectroscopy is often an issue for atmospheric retrievals. We can only wish that more funding is provided for improved spectroscopic measurements.

Table 3: The 'DIFF30' is a useful metric, it is given in %, but relative to what? Please be explicit. I'm actually surprised its values are so low (<25%) which is encouraging. Can you also indicate which sites are PROFFIT.

In the AMTD manuscript, the DIFF30 was given relative to the mean of the daily means for historical reasons (in previous work, the metric used was the standard deviation within a day). This explains why the % values did not correspond to the absolute values divided by the given mean (individual) TC (3rd column of Table 3). However, it is better to give this DIFF30 in percent relative to the mean of individual columns. Therefore, even if the numbers are very close, we have corrected the DIFF30 numbers in Table 3, and have specified in the legend to what they are relative. We have also changed the definition of our mean systematic error (7th column in AMTD): in AMTD version we did: mean(individual Syst / individual TC), and we now do: mean (individual Syst) / mean(individual TC). We then avoid too large effect of outliers or negative small columns.

Indeed, the DIFF30 values, which are, given the lifetime of HCHO of a few hours, an "empirical" measurement of the precision in our FTIR measurements, are quite low (median value of 9%) for a species with such very weak absorptions. We are also very pleased to reach such a good precision. The accuracy is less good (calculated as 14%), and this accuracy should be ideally also empirically evaluated by comparisons with correlative measurements.
The PROFFIT sites were already indicated with an * in Table 3 in AMTD. For AMT, we have changed this by ***, to be more clearly seen.

Page 13, lines 14-16. Some units are missing

Corrected for AMT version.

Page 13, last line. Variability faster than 30 mins. Is there any evidence for this in the literature (e.g., from models, campaigns).

We did not find any evidence for this tentative explanation from our side. We removed this sentence for AMT version, since it appears indeed too speculative.

Page 13, line 34: Typo – capital needed at ".this matrix..."
Page 15, line 7, Typo. "1E13" should be x1013

Corrected.

Figure 5: I can understand why this has been plotted but I think it would also probably be good to show the individual measurements for a single (common) year, rather than over the entire time record at each location. That way you can look at the day to day variability more closely – maybe put such a figure in the supplementary material.

We followed the Referee's suggestion. Instead of putting this plot of one single year in the supplementary material, we give it in Figure 5 in the new manuscript (chosen common year: 2016; except StDenis: 2011). and the complete time-series are given in supplementary material (Figure S1).

Figure 6. The variability in the HCHO observations poses some interesting questions. There is a lot of science in here.

Indeed. We hope that this data set will be soon exploited in modeling studies to explore the reasons for these different observed diurnal cycles. Some discussion and a few comparisons of FTIR and model diurnal cycles have been provided to answer Referee#3 's concerns about the diurnal cycle (but the model diurnal cycles are still not included in the revised manuscript).

Page 19. The 45% yield reduction – this maybe indeed correct – but can you provide a little more explanation/justification.

We have added the following sentence in the manuscript: “This fraction of 45% is higher, but of the same order, as the estimated overall impact of deposition on the average HCHO yield from isoprene oxidation (28%), based on IMAGES model calculations. The higher fraction for monoterpenes is intended to account for the impact of the more complex chemistry and larger number of oxygenated intermediates involved in their oxidation, compared to isoprene.”

Page 23: High mountain sites – at such locations it might be wise to quantify the difference between the station elevation and the model elevation for the 2x2.5 degree grid cells (maybe add information to Table 4). Is there any correlation between this difference and the bias?

Actually, the model takes the altitude of the station into account. We have added the sentence: "The model column is calculated from the calculated formaldehyde profile, between the altitude of the station and the model uppermost level (approximately 20 km), and from the a priori FTIR profile, above that level." Therefore, the overestimation of the model is related to the coarse resolution (2x2.5°), when the mountain site is not in clean area (e.g. Zugspitze, or Altzomoni which is in the same pixel than Mexico City), while the model performs well at mountain sites located in clean areas (e.g. Izaña or Maïdo for which the bias is the same as at StDenis located at the same island but close to sea level).

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