

Five years of CO, HCN, C₂H₆, C₂H₂, CH₃OH, HCOOH, and H₂CO total columns measured in the Canadian High Arctic

Anonymous Referee #2

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First, we thank the referee for these positive and insightful comments. We have responded to the comments and modified the paper accordingly as described below.

The manuscript "Five years of CO, HCN, C₂H₆, C₂H₂, CH₃OH, HCOOH, and H₂CO total columns" by C. Viatte et al. presents a very interesting new dataset of tropospheric trace gases that can be connected to biomass burning and anthropogenic pollution. Individually, all these gases have been measured before. However, measuring them all together and especially in the very sparsely covered Arctic is a significant scientific achievement. It also illustrates nicely the full potential of FTIR instruments for multispecies measurements – which is far beyond what other methods can provide. The manuscript is generally well written. However, some parts are somewhat lengthy. Especially the introduction could be shortened by 20-30%. The conclusions could also be more concise.

Specific comments:

- please shorten the introduction. 5 pages in AMTD format is a lot, 3 might also do.

We have shortened the introduction by cutting some detail. However, given the other referee's positive comments about the introduction, we have retained much of the literature review.

- the order of subsections in Section 2 is not logical. You should exchange subsections 2.2 with 2.3. This way, you would start with the description of the FTIR measurements in 2.1, continue with the discussion of microwindows in 2.2. After that, all the remaining subsections would be related to the OEM retrieval parameters.

As suggested, subsections 2.2 and 2.3 have been exchanged.

- p. 11361/2: calculation of the Sf matrix. Even though Batchelor et al. (2009) might have done it like that, I think it is not a good idea to make large perturbations in the 5% range to calculate Jacobians. In the linear case, it might not matter. However, even in a slightly non-linear case, 5% perturbation is a huge change. Partial derivatives should be calculated with infinitesimal changes!

We are currently testing the retrievals with the new SFIT4 code, which generates a full error analysis by calculating the Jacobians, the gain matrices, given the covariance-variance matrices for the retrieval parameters. This work is still ongoing, and the error budgets are of the same order of magnitude for the seven species.

- p. 11364, Equation (5): this equation is only valid if one measurement has a significantly higher vertical resolution than the other, so that its averaging kernel can be neglected (identity matrix). Is this the case for ACE-FTS? Otherwise, you would get a complicated convolution of both averaging kernels.

ACE-FTS uses the sun as a light source to record transmittance spectra during sunrise and sunset occultations through long atmospheric limb paths. Therefore, it has a vertical resolution of about 2–3 km in the troposphere (Tereszchuk et al., Atmos. Chem. Phys., 13, 4529–4541, doi:10.5194/acp-13-4529-2013, 2013), which is significantly higher than that of our FTIR measurements.

- Section 3.1: this section should be better structured. I would suggest to introduce two subsections: one discussing CO, C₂H₆, and C₂H₂ at the beginning and a second one discussing HCN, CH₃OH, HCOOH, and H₂CO which starts at p. 11366, l. 28.

We introduced two subsections in Section 3.1, one to discuss the seasonal variabilities of CO, C₂H₆ and C₂H₂ (subsection 3.1.1 related to the new Figure 10), and one for HCN, CH₃OH, HCOOH, and H₂CO (subsection 3.1.2 related to the new Figure 11).

- Section 3.1/Fig. 9: the measured (!) seasonal variabilities are the main science contribution from your work and should be given adequate space. You should split Fig. 9 into two independent figures: one for CO, C₂H₆, and C₂H₂ and another one for HCN, CH₃OH, HCOOH, and H₂CO). It might also be a good idea to somehow include the WACCM model results for the corresponding years. After all, WACCM is what you would have to rely on if you did not have the measurements. Therefore, difference between measurements and model results is the new information provided by the observations.

We have split Figure 9 into two separate Figures, one for CO, C₂H₆ and C₂H₂ and one for HCN, CH₃OH, HCOOH, and H₂CO, as suggested.

We decided to not show the WACCM model results here since the runs used to obtain the *a priori* VMR do not aim to reproduce the annual atmospheric variability, but to generate a representative average of the atmospheric state using self-generated winds for transport. However, we are currently comparing the FTIR measurements with model results from two chemical-transport models (GEOS-Chem and MOZART). The paper describing the comparisons between the PEARL FTIR dataset and the models is in preparation, and we plan to publish these results soon.

- Section 3.2: some of the correlations with ACE-FTS are not that good. You should discuss this more. According to Table 5, the two species with the smallest R have the highest number of coincident measurements.

We have inserted additional text discussing the different levels of agreement between the ACE-FTS and the PEARL FTIR datasets.

"The PEARL FTIR and ACE-FTS CO and C₂H₂ partial columns are in good agreement based on the slopes of the regression plots (FTIR vs. ACE-FTS). These values of 0.97 ± 0.11 and 1.21 ± 0.10

suggest no significant bias between the two CO datasets and a positive bias in the FTIR C₂H₂ relative to ACE-FTS. In contrast, the FTIR HCN and C₂H₆ partial columns are smaller than the ones measured by ACE-FTS, given the slopes of 0.69 ± 0.02 and 0.71 ± 0.04 , respectively. However, the FTIR HCOOH partial columns are significantly higher than those measured by ACE-FTS, with a slope of 3.35 ± 0.49 ."

- the conclusions are too lengthy. Please cut the summary part and focus on what you have learned from the measurements – e.g. with respect to what WACCM says about seasonal variability.

We have renamed the Conclusion section to "Summary and Conclusions" and cut the technical results to make it more concise.

Figures:

- Figure 1 is too messy and not very useful. It should be dropped.

We have decided to keep this figure but to split it into two separate ones, for greater clarity.

- Figure 9 is too busy. Please split according to the suggestions above.

We have split the Figure 9 into two separate ones, as explained above.