Atmospheric Measurement Techniques Discussions

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Comment on amt-2020-505

Chris Boone (Referee)

Referee comment on "First ground-based Fourier transform infrared (FTIR) spectrometer observations of HFC-23 at Rikubetsu, Japan, and Syowa Station, Antarctica" by Masanori Takeda et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2020-505-RC2, 2021

Ground-based column HFC-23 observations are reported for two locations, retrieved from FTIR measurements using SFIT4. Error sources are evaluated, trends are determined and compared to results from the AGAGE network. The results from the Japanese station exhibit large seasonal fluctuations that are attributed to transport from Asia, but the Antarctic results show no significant seasonal variation. The wintertime (December-January-February) results from the Japanese station are 10-15% lower than the AGAGE 12-box model estimates, while the results from the Antarctic station are roughly 25% lower than AGAGE estimates. The discrepancies are attributed in part to deficiencies in the temperature dependence of the pseudolines employed in the calculation of HFC-23, coupled with the latitudinal differences for the Antarctic station. Trend values for the two stations show generally good agreement with results from equivalent AGAGE measurement sites, supporting the use of ground-based FTIR measurements for HFC-23 trend monitoring.

Overall, the results appear to be of good quality, although I am not sure I agree with the interpretation of the seasonal HFC-23 variation (more on that later). The article is well organized, and the writing is clear, albeit with a few instances of wording/grammar issues.

I felt there was a missed opportunity here for placing the bias relative to the AGAGE results into a broader context. I agree with the authors' choice of using their winter (DJF) results as their "background level" to avoid the unexpected seasonal variation. On page 18, line 23, the authors state:

>…values from December to February are mostly stable with a relatively small standard deviations of about +/-10% and a value of 10–15% smaller than the AGAGE in-situ measurements of HFC-23.

Looking at Figure 4 of the Fernando et al article referenced in this paper, the global average results from the ACE-FTS exhibit very similar behavior, a persistent low bias relative to AGAGE results, roughly 10% in magnitude. Because the ACE-FTS employs the Harrison HFC-23 cross sections in its retrievals and the current study uses pseudolines based primarily on the same cross sections, this agreement in the bias relative to AGAGE does not seem like a coincidence. It looks like there is a systematic discrepancy between results derived using the Harrison cross sections and AGAGE results. I believe absolute calibration of the Harrison cross sections made use of the PNNL measurements, and I would be surprised if that group's determination of HFC-23 concentrations were off by 10% or so. However, I have no knowledge of the measurement technique employed by AGAGE, so I have no idea if a ~10% error on that end is feasible. It seems clear HFC-23 derived using the Harrison cross sections are inconsistent with AGAGE measurements, but I have no insight into the source of that discrepancy.

I am somewhat skeptical that the seasonal variation in measured HFC-23 is real. No other station sees such behavior. They theorize that it comes from long range transport, and yet not a hint of it continues on across the Pacific Ocean to Trinidad Head, despite the apparent large magnitude of the emissions. It is possible the station in question is located in just the right location downwind of a large emitter, and perhaps the flow would routinely divert away from Trinidad head as it travels across the Pacific (not my area of expertise). Emissions during HCFC-22 production is a major source of HFC-23, and Japan is a significant HCFC-22 producer, so perhaps the emissions originate from near the station, which would explain why they see such large quantities. However, I would be inclined to think there might be an artifact in the retrieval. Are there systematic differences in the residuals for baseline HFC-23 conditions versus enhanced HFC-23 conditions? If additional structure appears in the residuals in the latter case, the retrieval may be compensating for something missing in the analysis or compensating for large residuals from some other constituent (e.g., from water lines if H2O levels are high). Is there a strong correlation of enhanced HFC-23 with H2O, temperature, the ratio of HDO to H2O, seasonal variations in CH4, or biomass burning products like HCN or C2H6? I suspect H2O might be the most dangerous of the bunch for impacting the retrieval. Correlation does not prove cause but may help identify the source of a retrieval artifact, if one exists.

In my opinion, asserting that the HFC-23 enhancements were real would at minimum require comparing residuals for enhanced versus background conditions, ideally with a similar quality in both cases and no evidence of additional significant systematic features in the residuals for the enhanced case.

The authors use two microwindows in the retrieval. I assume independent 'background correction' parameters (slope and curvature) are used in each window. Is it possible to have SFIT4 use a single set of background correction parameters spanning the two windows? The windows are close together, so I expect a unified curve for the background correction should be viable. This adjustment would presumably make the retrieval less susceptible to artifacts.

Page 8, line 25: for all observed spectra with the IFS-120/5HR instrument no ILS function was used because the instrument has always been maintained best optical alignment

This statement is not entirely accurate. An instrumental line shape function is still required, associated with the FTS scan length and the finite field of view. It would be more appropriate to say something like the ILS is accurately defined by the theoretical model for the given instrument configuration.

Page 4, line 9: However, the ACE-FTS observations do not have sensitivity to the troposphere where all HFC-23 emissions occur.

Also not entirely accurate as stated. ACE-FTS measurements extend into the troposphere (i.e., below the tropopause). It would be more accurate to say ACE-FTS measurements do not extend low into the troposphere or do not probe near the surface.

Page 20, line 3: a discussion of temperature sensitivity in relation to the ACE-FTS.

Note that ACE-FTS analysis uses the cross sections directly in the analysis (via a bilinear interpolation in pressure and temperature), whereas the current study employs pseudolines derived from a set of cross sections. I would assume that the deficiencies in temperature dependence described in the paper are a property of the pseudolines and not the cross sections themselves. There could conceivably be temperature consistency issues arising from the ACE-FTS interpolation approach, but I see no reason to expect it is

similar in nature to the pseudoline temperature dependence deficiencies unless one claims the temperature dependence problems are inherent to the Harrison cross sections (I would need to see proof , if that were claimed).

Page 7, line 25: only about 1% of the atmospheric transmittance of solar infrared radiation at ground level (see Figure 3).

It is traditional to make reference to figures in order. Figure 3 is referenced before Figures 1 and 2.

Page 17, line 21: ð□□□ is the column-averaged acceleration

acceleration due to gravity

Grammar/wording issues:

>Page 3, line 6: all UNFCCC CDM project were terminated … [projects]

>Page 5, line 31: detected with the MCT detector … [measured with]

>Page 5, line 31: Note, that … [no comma]

>Page 11, line 31: DOFS for HDO was increased when the wider window used … [was used]

>Page 14, line 7: due to the lack of vertically measurement data … [vertically resolved]

>Page 14, line 31: The SZA random uncertainty was assumed an uncertainty of 0.15º … [assumed a value of]

>Page 15, line 18: Since the MW for CH4 pre-retrieval is closed to the HFC-23 MWs … [close to]

>Page 16, line 29: These relative biases lead underestimation to the trend … [lead to an underestimation of the trend]

>Page 19, line 32: in all temperature region … [regions]

In conclusion, while I think it is a good paper, I would like to see more said on AGAGE results and results from studies employing the Harrison cross sections (this study and ACE-FTS work) exhibiting a fairly consistent bias, suggesting a systematic error somewhere. I would also like to see further proof that the apparent seasonal enhancement in HFC-23 is real (e.g., comparing residuals in the enhanced case versus the background case).