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Atmospheric Measurement Techniques Discussions

Interactive comment on "The water vapor self-continuum absorption in the infrared atmospheric windows: New laser measurements near 3.3 μm and 2.0 μm" *by* Loic Lechevallier et al.

Anonymous Referee #2

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The manuscript is devoted to measurements of the water vapor self-continuum in the two near-infrared spectral regions. Additional experimental data is presented to those already reported by the same group for other spectral regions. The paper is clearly written, easy to read, and present rather important information about water vapor self-continuum absorption in narrow 3-5 mm radius cells in equilibrium conditions. I think this is a good paper to be published in AMT after accounting for the comments described below.

Main issues:

1. The authors often write about the atmospheric application of their continuum mea-

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Discussion paper

surements. However, all CRDS measurements by Grenoble group have so far been performed in very narrow (3-5 mm radius) cells where conditions for water vapor can be rather different from those in the atmosphere or in much wider FTS cells. I would not raise this issue for many other gases, but water vapor is a specific case. It is interesting that there is rather close agreement between different measurements of the foreign water vapor continuum, but for the self-continuum, the situation is quite different. Different measurements performed applying different methods give very different results up to more than an order of magnitude.

There is some available evidence, including from satellite measurements (see, for example, the Anonymous Referee #1 comment: “The Richard et al. value was considered in developing recent MT_CKD version, but found to not be consistent with the satellite- and ground-based observations analyzed in Mlawer et al. (2012)”), to suggest that the water vapor self-continuum absorption in the atmosphere can be much stronger than that obtained from 3–5 mm cells in CRDS measurements. One may suggest that water vapor at low pressures may be, for example, depleted of long-living stable water dimers (with a lifetime >= 0.001 s) and/or water nanoclasters in such narrow cells due to their adsorption on cell walls. So, the OFCEAS/CRDS continuum data may be correct for a monomolecular gas, but underestimates atmospheric continuum absorption. I suggest that the authors explicitly recognize this issue in their revision both in their introduction and conclusions.

2. Page 9, lines 14-17: “the FTS values are largely overestimated compared to both MT_CKD and the laser measurements. Compared to OFCEAS, CAVIAR and Tomsk 2015 CS values, reported with a 50 % error bar, are overestimated by a factor of three while the FTS values reported by Baranov and Lafferty with a 22% error bar are overestimated by a factor of two”.

This is a somewhat misleading statement. The authors do not take into account the rather strong T-dependence of the self-continuum. They should keep in mind that the OFCEAS data which they show corresponds to about 300 K, while Baranov&Lafferty

data are shown for 311 K, and Tomsk-2015 data, for 287 K. To extrapolate to 300 K, the Tomsk data should be decreased by ~30%, while Baranov&Lafferty data should be increased by ~25%.

The T-dependence detected in Baranov & Lafferty's measurements is even stronger than that (see Table 2 in Baranov & Lafferty, JQSRT, 2011 or the left panel of Fig. 7 in the current manuscript). In fact, the Baranov&Lafferty data show a more consistent T-dependence in this window than the present OFCEAS continuum data, and a T-dependence which is as consistent, if not more consistent, with independent FTS data in this same region. So, I personally am not sure which, of OFCEAS or B&L's data, I should trust in this window. The wording here also needs to be more careful, to make clear that is it is specific to near room-temperature.

3. Page 16, lines 5-15: The authors' comments are somewhat misleading in this paragraph, and the authors seem to too easily neglect the results of the FTS measurements. The 0.1–0.2% is a *prediction* of the CRDS measured absorption to the FTS measurement conditions and the argument could be characterized as "circular" – it uses the CRDS measurements to establish that the CRDS measurements must be most reliable! However, the level of continuum absorption measured in FTS experiments in 4 mcr window at close-to-room temperatures was not 0.2%, but 1-1.5 % in Tomsk (2013) and 2–3% in Baranov & Lafferty (2011) measurements. These are rather high values and cannot simply be neglected or characterized just as an "error" (Referee #1 comment). Baseline instability was thoroughly investigated both from pure nitrogen and empty cell absorption before and after the sample measurements, and - at least in Baranov & Lafferty measurements - was several times smaller than the measured continuum absorption:

"Periodic and multiple tests were made to establish the baseline stability. In recording spectra with the cell filled with pure nitrogen up to about 400 kPa (4 atm), no significant systematic changes in baseline caused by mechanical changes in the cell were observed. With much smaller water-vapor pressures, only a small random scatter within

0.3% (one STDV) was observed. A slow drift of the baseline in time was eliminated by averaging spectra of the empty cell recorded before and after the sample spectrum" /Baranov & Lafferty, JQSRT, 112, 1304-13, 2011/

FTS measurements at different pressures were also performed by Baranov & Lafferty (JQSRT, 2011) in contradiction to the implication at lines 11 to 13 of the manuscript.

Again in this paragraph a general statement is made about the quality of FTS measurements without specifically clarifying that the argument is only valid at room temperature as is clear from Figure 7 (and there is evidence, discussed above, that the argument may not even valid at room temperature in atmospheric conditions).

Recent FTS measurements in Tomsk, reported at HRMS-2017, 20–25 August, Helsinki, Finland, were performed at several water vapour pressures at 316 K and a path length of 1000 m, and have detected continuum absorption from 4 to 10% with pressure-squared dependence. They agreed well with Baranov & Lafferty's result for 2500 cm-1 window.

Therefore, I have serious doubts that self-continuum absorption measured in 3-5 mm radius CRDS or OFCEAS cells can be simply applied to atmospheric conditions before the real reasons for such strong disagreement between CRDS/OFCEAS and FTS room-temperature measurements in windows is clarified.

Minor issues:

"Self-continuum" and "Self continuum" – should be unified in the manuscript.

Page 1, line 18: "completing" is exaggerated. First, the 4 micron window is only covered by 3 measurements and at all wavenumbers the range of temperatures measured, compared to what is needed for accurate atmospheric measurements, is sparse. The authors acknowledge this more correctly at p.17 (10).

Page 8, line 6: "on the order of 30 %." According to the Table 1, it is rather about 25%"

Page 8, line 10: “water continuum represents about 25 % of the measured signal (Fig. 2)”. According to Fig. 2 it is rather 27-30%.

Figs. 2 & 5: “The contribution of water monomer” is better written as “The simulated contribution of water monomer”.

Page 9, Fig.3: Temperatures should be given for OFCEAS measurements and MT_CKD models. This is particularly important given that the OFCEAS measurements vary significantly over a small temperature interval in Fig. 7, and the degree of agreement with MTCKD at 298 K is not found at all at 303 K.

Page 12, line 13: “of the calculated WML contribution calculated as indicated above” – remove the repeated “calculated”.

Page 12, line 14: “the WML contribution represents between 18 and 38 % of the total absorption (see Fig. 5)” Again, according to Fig. 5 it is a rather firm 35–38% (nearly independent on pressure). Why do the authors give such uncertain values?

Page 14, Fig 7. The measurements in the 3000 (more properly 3007 cm-1) window do not seem to agree with Table 1. In Table 1, only one point (at 303 K) is over 2.0, and yet 2 points in Fig 7 exceed this value. Might the difference between neighboring points also indicate that the authors are too optimistic in their uncertainty estimates?

Page 17, line18: See also 10.1029/2007GL029259 for evidence of water dimers in the region under consideration in this paper.

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