Author Response, Andreas Reichert, Karlsruhe Institute of Technology, Garmisch-Partenkirchen, Germany, 30 August 2016

Dear Dr. Maring,

a point-by-point response to the reviews to our manuscript acp-2016-323 is given below and available online at http://www.atmos-chem-phys-discuss.net/acp-2016-323/acp-2016-323-AC1-supplement.pdf. We thank for the sound and very constructive referee and short comments that greatly helped to improve our manuscript. A marked-up version of the manuscript highlighting all changes made by the authors is also attached to this response.

We are confident that all referee comments have been addressed thoroughly and hope that a final publication in ACP is possible soon.

Sincerely,

Andreas Reichert, 30 August 2016

Author response to the referee and short comments:

We thank the referees and K. P. Shine et al. for their very sound, constructive and helpful comments which helped us to significantly improve our manuscript. In the following, we provide point-to-point replies to all comments made by the referees. All page and line numbers quoted in this reply refer to the initial version of the manuscript.

Anonymous Referee #1

Comments :

In Supplementary material the authors give mean water vapor continuum absorption coefficients with negative values which have no physical meaning. Moreover data are given with large uncertainty in the center of the windows so that they are in agreement with all the literature data within the error bars bringing no additional information for those spectral regions. They also weaken the other data obtained in the bands and at the edges of the windows with lower uncertainty. To me it will have more sense to remove large uncertainty data in the center of the 2.1 μm, 1.6 μm windows before publication.
As mentioned in the companion paper Part 1: Very dry atmospheric conditions are a pre-requisite for closure studies of this kind due to the otherwise saturated spectral regions. This has for consequences that in these conditions of dry air, the foreign-continuum represents most of the continuum absorption (more than 70% if the MT_CKD self to foreign ratio is assumed according to the authors).
This can be viewed as a kind of “limitation” of the method but it is also important as there is a real lack of observational constraints for the foreign-continuum.
To conclude, this paper presents state-of the art atmospheric measurements of the continuum which bring interesting information in the near infrared bands and at the edges of the windows mostly for the foreign-continuum for which experimental constraint are clearly

missing. For these reasons this paper deserves to be published in ACP after large uncertainty data in the center of the 2.1 µm and 1.6 µm windows are removed.

We agree with the referee that in many window regions, our results have too large uncertainties to be useful for comparison to other studies or follow-up analysis. As suggested, we therefore removed data points for which the estimated errors exceed the measured continuum absorption from Fig. 1.
However, we respectfully disagree on the statement that the removed measurements have no physical meaning. The mean absorption coefficients have negative values in many window regions, which indicate systematic errors e.g. in AOD measurements or radiometric calibration. However, these possible errors are included in the uncertainty budget and within the given uncertainties, our results are consistent with a positive continuum and with recent continuum quantification studies in these spectral regions. We think that the results may still be valuable since they provide an upper limit to the continuum and therefore included the full results in the revised manuscript the form of an appendix.

Specific comments

P1, L32: Burch (1982) and Burch and Alt (1984) used a grating spectrometer for their experiments and not a FTIR spectrometer.

We thank for pointing out his wording mistake. The manuscript was corrected (Page 1, line 38): "Several studies made use of cell measurements with grating spectrometers (e.g. Burch 1982; 1985; Burch and Alt 1984) or FTIR (Fourier Transform Infrared) spectrometers..."

The following references are missing for continuum measured:
By CRDS:
- Mondelain D. et al., J. Quant. Spectrosc. Radiat. Transfer, 130, 381 (2013).
- Cormier, J. G. et al., J. Chem. Phys., 122, 114309, (2005)
- Cormier J. G., et al., J. Chem. Phys. 116,1030 (2002)
By OF-CEAS:
- Ventrillard et al. J. Chem. Phys. 143, 134304 (2015)
By calorimetric-interferometry :
- Fulghum, S. F., and M. M. Tilleman, J. Opt. Soc. Am. B Opt. Phys., 8, 2401(1991)

The additional references were added to the manuscript as suggested by the referee.

P1, L38: The sentence is too general. Cavity enhanced techniques like CRDS and OF-CEAS as well as CI are able to measure continuum absorption in the windows at room temperature (see references above) and even at lower temperature (see Cormier 2005).

The wording in the manuscript was changed to avoid misrepresentation (Page 1, line 38):
"A further challenge for laboratory studies is that they are typically carried out at higher temperatures than those encountered in the atmosphere in order to detect the weak continuum absorption in the limited optical path length of the cells. Note that CDRS and related techniques in principle enable measurements at atmospheric temperature (see e.g. Cormier et al., 2005) but such measurements are not yet available for many spectral regions."

P2, L1-3: The sentences "To date…non-straightforward" are too definitive. Temperature dependences of the self-continuum cross-section have been investigated in different spectral windows (see for example Cormier 2005, Mondelain 2014, Ptashnik 2011, Ventrillard 2015…). In the 2.1 µm window, for example, the temperature dependence measured at high

temperatures by the CAVIAR consortium is similar to that measured at lower temperature by Ventrillard et al. in different part of the window. For sure there is a real lack of observational constraints for the foreign continuum.

The manuscript was changed as follows to avoid the misleading statement in the initial manuscript (Page 2, line 1):" To date, the temperature dependence of the self-continuum has been investigated by measurements in a number of spectral regions (e.g. Cormier et al., 2005; Mondelain et al., 2014; Ptashnik et al., 2011; Ventrillard et al., 2015). However, the remaining uncertainty of the self-continuum temperature dependence (see e.g. Paynter and Ramaswamy, 2011) and the lack of measurements of the foreign continuum temperature dependence cause considerable uncertainties in the application of the laboratory results on atmospheric radiative transfer calculations."

P2, L4-5: To me it is much more difficult to characterize the continuum from atmospheric spectra than from laboratory spectra recorded in well-known conditions of temperature with sufficiently sensitive and stable techniques. In atmospheric conditions additional uncertainties occur due to water vapor profile, temperature profile, aerosols uncertainties and it is more difficult to separate self and foreign continuum.

The following discussion was added to the manuscript (Page 4, line 8):" While such atmospheric closure studies enable to avoid some limitations of laboratory measurements as outlined above, they are also subject to a number of major challenges: absorption in the NIR due to aerosols can become comparable to the magnitude of the water vapor continuum absorption of interest (Ptashnik et al., 2015) and requires an accurate separation of continuum and aerosol contribution. Furthermore, the characterization of the atmospheric state (e.g. IWV, water vapor profile, temperature profile, and further trace gas column amounts) is more challenging and typically less accurate than the characterization of experimental conditions in a laboratory study."

P2, L23: Which version of the HITRAN database is used for the line-by-line calculations? Is the line profile used is a Voigt profile truncated at +/- 25 cm-1 from the line center with the plinth subtracted? If yes the authors have to mention it.

The following text was added to the manuscript (Page 2, line 23): "Spectral line parameters were set according to the aer_v3.2 line list provided alongside the LBLRTM model."
Page 2, line 24: "We adopt the definition of the water vapor continuum given in Turner et al. (2010), i.e. water vapor continuum is defined as all absorption by water vapor exceeding a Voight line shape within ± 25 cm-1 of each line center minus the value of the Voight line shape at ± 25 cm-1 ("plinth")."

P7, L32: How exactly calculations are done in the case of Bicknell et al. (2006) as these measurements did not allow dissociating the self from foreign continuum.

The following text was added to the manuscript (Page 7, line 32): "Since the results of Bicknell et al. (2006) do not allow a dissociation of self from foreign continuum is not possible, we assumed the self-to-foreign ration suggested by the MT_CKD model to calculate the corresponding value of $\overline{\boldsymbol{k}}_{cont}$. "

P7, L8: Assuming that the partitioning in self and foreign continuum given by the MT_CKD model is a quite strong hypothesis as the very few laboratory studies of the foreign-continuum (Ptashnik PTRSA 2012 and Mondelain PCCP 2015) seem to show that the foreign continuum cross-sections are largely underestimated by MT_CKD in the windows.

The following text was added to the manuscript (Page 7, line 8): "Note that this assumption has to be considered tentative since for both self- and foreign continuum the results of recent laboratory studies deviate from the MT_CKD model especially in window regions (e.g. Ptashnik et al, 2012, 2013; Mondelain et al., 2015)."

P7, L38: The very good agreement with CRDS-based measurements of Mondelain et al. (2015) is essentially a good agreement with the foreign-value measured in this paper due to the dominating contribution of the foreign-continuum in the atmospheric conditions encountered at Zugspitze. This is an important point as in the Mondelain et al study the measured foreign-cross section is 4.5 times larger than the one given in MT_CKD 2.5. The fact that in the 4100 to 4200 cm^{-1} spectral range the MT_CKD model underestimate the continuum goes in the same direction as well as the Ptashnik 2012 paper. This point should be underlined by the authors.

The following discussion was added to the manuscript (Page 7, line 38):"Due to the dominant role of the foreign continuum in the 4100 to 4200 cm^{-1} spectral range, this agreement mainly corresponds to a comparison of the foreign continuum results of Mondelain et al. (2015) and our measurements. Therefore, our results are consistent with the finding of Mondelain et al. (2015) and Ptashnik et al. (2012) that the foreign continuum is underestimated by the MT_CKD model in this spectral region."

Fig 1: Some literature data at room temperature are not plotted on this figure (Ventrillard 2015, Mondelain 2014). It will be good to incorporate them in the figure.

The intention of Fig. 2 (previously Fig. 1) was to present a comparison of our results to measurements which comprise a quantification of the self and foreign continuum at or below room temperature using the same (as e.g. for Mondelain et al., 2015) or a very similar experimental setup (as for Ptashnik et al. 2012, 2013).
To clarify this intention, the following text was added to the manuscript (Page 6, line 30):
"The figure includes laboratory measurements carried out at or below room temperature which provided constraints on both the self and foreign continuum using the same or a very similar experimental setup."
The measurements of Ventrillard et al. 2015 and Mondelain et al., 2014 were not included in the figure since they only comprise results for the self continuum, while additional assumptions have to be made on the foreign continuum to calculate the overall continuum absorption. However, we agree on the relevance of these studies and have therefore referenced and discussed them in the manuscript.

Technical corrections:

P2, L25: closure experiment und the related uncertainties: und->and.

The manuscript was changed as suggested

Referee #2, Penny Rowe

Main comments:

1) Literature review. I do not think it is necessary to exhaustively reference laboratory work performed over a different spectral range. But all lab work that overlaps the spectral region should be referenced. (Thus I think some, but not all, of the references suggested by the

other reviewer need to be included). In addition, studies in atmospheric conditions with similar instruments should be referenced (although they are mainly in different spectral regions):

Page 2, line 5. Please include the following references with a sentence such as, "The continuum has been investigated for atmospheric conditions using measurements of atmospheric emitted infrared radiance for other spectral regions (Tobin et al., 1999; Rowe and Walden, 2009) and for part of the region of interest for this study (Newman et al. 2011; 2400 to 3200 cm-1), but not for the spectral region 3200 to 7800 cm-1."
- Tobin et al.: Downwelling spectral radiance observations at the SHEBA ice station: Water vapor continuum measurements from 17 to 26 micron, J. Geophys. Res., 104, 2081-2092, 1999.
- Rowe, P.M. and Walden, V.P.: Improved measurements of the foreign-broadened continuum of water vapor in the 6.3 micron band at -30 C, Appl. Opt. 48, 1358-1365, 2009.
- Newman et al. 2012: Airborne and satellite remote sensing of the mid-infrared water vapour continuum, Phil. Trans. R. Soc. A 370, 2611-2636.

*The following literature review was added to the manuscript as suggested by the referee (**page 2, line 5**):*
"The continuum has been investigated for atmospheric conditions using measurements of atmospheric emitted infrared radiance for other spectral regions (e.g. Tobin et al., 1999; Rowe and Walden, 2009). However, atmospheric measurements are available only for a fraction of the spectral region covered by this study (Newman et al. 2011; 2400 to 3200 cm^{-1}), while for the remaining interval from 3200 to 7800 cm^{-1}, no atmospheric measurements have been reported."

The results of Newman et al. should also be discussed (e.g. on Page 7, line 38).

*The following discussion was added to the manuscript (**page 7, line 38**):*
"A fraction of the spectral range covered by this study, namely 2500 -3200 cm^{-1}, was also included in the airborne measurements by Newman et al. (2011). Newman et al. (2011) conclude that the increase of the self continuum in MT_CKD 2.5 compared to MT_CKD 2.4 lead to reduced spectral residuals, while no firm conclusion can be drawn in the 2500 - 3200 cm^{-1} –range on whether MT_CKD 2.5 or the results of Ptashnik et al. (2011) represent a more appropriate quantitative description of the water vapor self continuum. These findings are in agreement to the results of this study, given that both are not consistent with continuum absorption being weaker than indicated by MT_CKD 2.5. "

Also on **Page 8, line 20**, change "in this spectral range" to "for most of this spectral range."

The manuscript was changed as suggested.

2) Fig. 1. If you also include kcont for the self and foreign-broadened parts of the MT-CKD continuum separately in this plot, it will show the relative importance of each for your results. If any of the lower bounds on your error bars are significantly different from zero, you might make those error bars black instead of gray so they stand out more. In the caption, "(black)" needs to be changed to "(black; gray error bars are shown for points for which only the upper threshold can be determined to within the uncertainty)." If you have measurements that are not shown on this plot, you should create a second panel below on a linear y-scale where they can be seen. In the caption, state something like: "x points that fall outside the plot region are not shown in the upper panel (log scale) but are evident in the lower panel (linear scale)"

The contributions from self- and foreign broadened MT_CKD-continuum were included in Fig. 1 as suggested. Only data points with significant continuum absorption were included in Fig. 1 (as suggested by referee #1), while the entire set of results is shown in the supplementary figure Fig. B1

3) (Optional) The paper would likely receive more citations if the following changes were made.

- Title: Stating the spectral range explicitly in the title will help readers more quickly determine if the paper is of interest to them, especially given the large spectral range. (You could omit "under atmospheric conditions" because the title already includes "radiative closure experiment.")

The title was changed to: "Quantification of the mid- and near-infrared water vapor continuum in the 2500 to 7800 cm^{-1} spectral range under atmospheric conditions"

- Cf. If you estimate the foreign-broadened part of the continuum (Cf), it can be compared to previous work, and incorporated (e.g. in figures) in future publications by other authors. I suggest estimating the self-broadened continuum (Cs) based on what you think is most accurate (MT-CKD or other; give rationale) and the atmospheric water and temperature structure, removing its effects from k_{cont}, and calculating Cf. Increase your error bars correspondingly (uncertainty estimate for self-broadened continuum x 0.1 to 0.3). Discuss how you calculated Cf briefly in the text. Add a figure showing the subset of results for which the error bars are small enough to be useful.

The following text was added to the manuscript (Page 7, line 12):
"If values for c_f are required, further assumptions on the self continuum have to be made before subtracting this contribution. As an example, Supplement B to this manuscript contains a list of c_f-values for all spectral bins where c_f exceeds the uncertainty estimate. The results were calculated from our measurements assuming the self continuum to be consistent with the MT_CKD model. Recent laboratory measurements (e.g. Ptashnik et al., 2013) suggest that this assumption may not be appropriate. However, alternative sources of the self continuum neither constitute a more robust estimate, given the inconsistencies between different laboratory results, the uncertainty of the self continuum temperature dependence and the fact that the foreign continuum is likely to be the dominant contribution to the overall continuum absorption for the dry atmospheric conditions of our study and the spectral windows covered by the measurements (see Fig. 1). A 50 % uncertainty was assumed for the self continuum as suggested by Paynter et al. (2011) and is included in the uncertainty of c_f in addition to the uncertainty budget presented in Sect. 3.2."

Minor comments

- Give the wavenumber range the first time you mention each region (near-infrared, etc)

*The wavenumber ranges were added to the manuscript. **Page 1, line 23**: NIR, "4000-14000 cm^{-1}", **page 2, line 11**: "FIR, 2-667 cm^{-1}", **ib**. "MIR, 667-4000 cm^{-1}"*

- **Page 4, Line 8**: Please show examples of measured and synthetic radiance spectra. (You can alternatively reference your other paper here, but I think it would be nice to have it here as well).

*The following text was added to the manuscript (**page 4, line 9**):*
"Figure 1 shows the mean measured and synthetic radiance spectra for the closure data set that will be presented in Sect. 3.3."

A figure showing the mean measured and synthetic radiance spectra was added to the manuscript (Fig. 1):

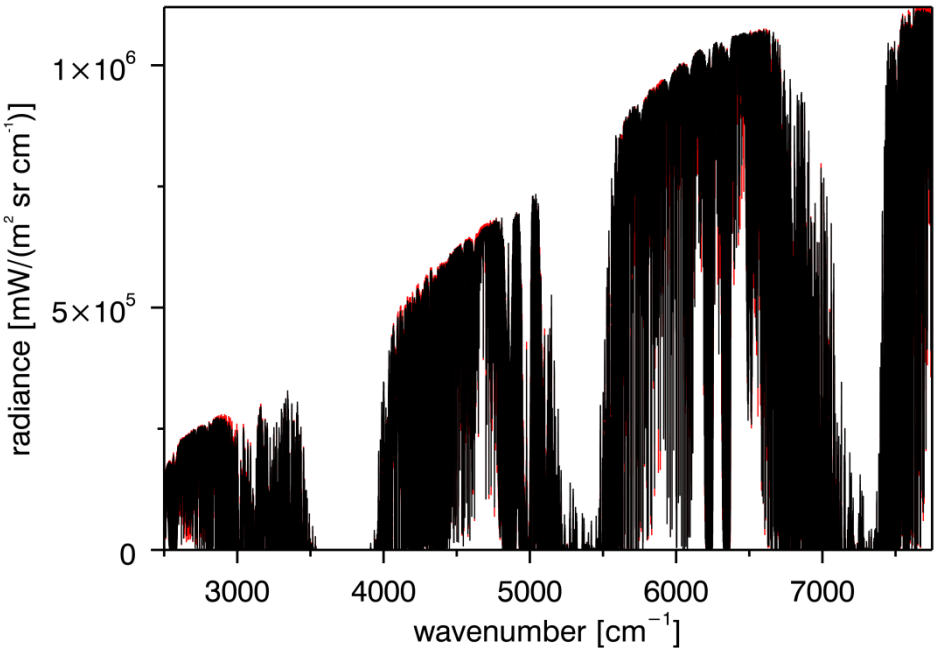

Fig. 1: Mean measured (black) and synthetic (red) radiance spectra for the closure data set selected according to the criteria presented in Sect. 3.3.

- **Page 4, line 20 (approx.)**. You might mention here that cs is strongly temperature dependent but that cf is thought to be only weakly temperature dependent.

*The following text was added to the manuscript (**page 4, line 26**):*
"In addition to their different dependence on water vapor density according to Eq. 5, self- and foreign-broadened continua are characterized by their distinct temperature dependence: while the self continuum shows strong negative temperature dependence, the foreign continuum is assumed to have no or only weak temperature dependence."

Technical and grammar corrections

- **Page 1, Line 26**, remove the word "exact"
- **Page 1, Line 31**, change "both continuum" to "continuum absorption, including the contributions of both the self and foreign-broadened continuum"
- **Page 2, Line 10**, change "thereafter" to "hereafter"
- **Page 3, Line 4**, add the word "for" before "data"
- **Page 3, line 8**, change "disposes" to "consists of" or "includes"
- **Page 3, line 9**, rephrase "centered at nm." Perhaps give the range of the channels or the bounds of each.

***Page 3, line 9** was rephrased as follows: "Only information from 5 channels whose central wavelengths are in the spectral region between 439.6 and 781.1 nm was used in the analysis. The exact filter wavelengths and full width at half maximum (FWHM) values of these channels are listed in Table 1."*

- Page 3, line 24, Do you mean errors in the AOD measurements from the sun photometer (rather than errors in the sun photometer measurements)? If so, add "AOD

determined from" before "sun photometer measurements."

"AOD determined from" was added on **page 3, line 24**

- **Page 3, lines 26-27**, "The following measurements." The sentence is awkward, rephrase.

*The sentence was rephrased as follows **(page 3, line 24):** "The AOD uncertainty comprises several contributions: First of all, the AOD determined from the sun photometer measurements…"*

- **Page 4, line 11**, change "the criteria presented" to "criteria that will be presented"
- **Page 6, line 6**, change "requested" to "required"
- **Page 7, line 21**, change "where treated in sufficiently" to "were treated in a sufficiently"
- **Page 8, line 18**, change "presented" to "present"
- **Page 9, line 12**, change "we thank for support by the" to "we are grateful for support by the"
-**Table A1**. Convert into two tables, putting further parameters in a separate table.

All technical and grammar corrections were applied to the manuscript as suggested by the referee.

Short comment by Shine et al.

Reichert and Sussmann (2016) present an important attempt to characterise the water vapour continuum in the near-infrared in atmospheric conditions. Given that relatively few such measurements exist, such work is very welcome.

We have a number of comments on the paper. The major one relates to our comment on Part II of this paper, where the authors calibrate their measurements to an assumed extraterrestrial solar spectrum (ESS); as we note in that comment, there are significant uncertainties in the ESS. This uncertainty has important consequences for the derivation of the continuum, especially in the window regions, which are not taken into account here.
It is our view that this uncertainty renders the continuum derivations here unreliable in window regions; the fact that many of the derived continuum values in the windows are negative and therefore unphysical (as shown in the data in their Supplement but not in the figure in the paper) adds support to the opinion given by Reviewer 1 (10.5194/acp-2016-323-RC1) that the derived continuum values deep in the window are so uncertain that they should not be presented.

Major comments

1. Equations (2) and (3) derive the continuum optical depth from the difference between the observed downward radiance at the surface and the modelled radiance ignoring the continuum. To do this reliably requires that the ESS is well constrained. This is not currently the case, as we explain in our comment in Part II (see e.g. Thuillier et al. (2015) and Weber (2015)).
Various derivations from satellite and other observations differ by 5 - 10%. The authors' method is essentially to write a radiance residual (their Equation (2)) between observations and model so that

$$\Delta I = S_{actual} \exp(-(\tau_g + \tau_{cont} + \tau_{aer})) - S_{model} \exp(-(\tau_g + \tau_{aer}))$$

where τ is the optical depth due to lines of the gases (subscript g), water vapour continuum (cont) and aerosols (aer), and S_{actual} and S_{model} are the actual ESS and the ESS used in the model respectively.
Since S_{actual} is not observe, the authors (in Part II of the paper) perform a Langley analysis on their observations to derive $S_{Langley}$, and then apply a calibration constant (c) to force $S_{Langley}$ to agree with S_{model} (i.e. $S_{model} = cS_{Langley}$). The authors note in Part II that their "closure validation does not provide information on the accuracy of the used ESS" but here we are concerned about the impact of this on the radiance residual.

τ_{cont} is then derived from the above equation as

$$\tau_{cont} = -\ln\left(\frac{S_{model}}{S_{actual}}\left(\frac{\Delta I}{S_{model}\exp\left(-(\tau_g+\tau_{aer})\right)}+1\right)\right).$$

If $S_{model} = S_{actual}$ (i.e. if S_{model} is indeed the true value), then this equation reduces to the authors' Equation (3). However, if this is not the case, then any error in the ESS (which would lead to a radiance residual even if τ_{cont} is zero) gets incorrectly attributed to τ_{cont} – the resulting error in τ_{cont} is particularly severe for the low values of optical depth found in the window regions, and even the sign of τ_{cont} is not constrained to be positive.
We believe that it is important to incorporate the effect of errors/uncertainties in the assumed ESS. We expect that such an analysis will lead to the conclusion that the derived values of the continuum in the centres of the windows are too unreliable to be presented.

We thank K. P. Shine et al. for pointing out recent research that indicates that the ESS uncertainty may be significantly higher than assumed in our initial manuscript. An extensive discussion of recent ESS results has been included the companion publication Part II which introduces the radiometric calibration method used in our analysis.
In their comment, Shine et al. raise the question whether the continuum results might be significantly influenced by inaccuracies in the used ESS. A discussion of this important point was missing in the initial version of the manuscript, and we thank Shine et al. for highlighting this issue.
In their first equation, Shine et al. make a definition of the spectral residual ΔI. Based on this definition, they conclude that errors in the ESS might have significant influence on the determined continuum OD.
We agree that the analysis made by Shine et al. is correct and their first equation represents a valid definition of the spectral residual. However, the definition of spectral residuals made in Eq. 2 of our manuscript, which corresponds to the continuum quantification procedure in our study, is not fully equivalent to the definition given by Shine et al.
While the first term of the definition of Shine et al. contains the 'real' downwelling solar radiance (i.e. without ESS errors), our definition relies on $\boldsymbol{I}_{FTIR}$, i.e. the measured downwelling radiance. The measured radiance according to Eq. 2 of our manuscript already contains ESS errors due to the use of the ESS in the radiative calibration. Note that both the modeled and the measured radiance rely on the same ESS, which as noted rightly by Shine et al. may be prone to substantial inaccuracies. The design of the continuum quantification procedure used in our study thereby greatly reduced the impact of ESS errors on the continuum results.

The influence of ESS errors on the continuum results for the continuum quantification method presented in our manuscript can be described as follows:

a) For spectral points where only Langley data was used for the calibration, ESS errors have no influence on the continuum results. The situation at these points is equivalent to the well-known Langley calibration of sun photometers where no information on the solar spectrum is needed to infer accurate atmospheric optical depth.
b) In between the Langley points, where additional blackbody measurements are used for calibration, there is in fact a second-order influence of ESS errors on our continuum results. To investigate the magnitude of this effect, we repeated the continuum quantification analysis presented in the manuscript using the high-resolution ESS by Menang et al. (2013) instead

of the ESS by Kurucz (2005). Apart from the exchange of the ESS, the data set and analysis was not modified. This is a good test to assess the sensitivity of the results to ESS uncertainty since the ESS by Menang et al. (2013) differs from the Kurucz-ESS by about 5 % (recent ESS results generally feature differences of up to ±5 % compared to the Kurucz-ESS) and features many solar lines not included in the Kurucz-ESS. Note that the Menang-ESS covers the spectral range $\nu > 4000\ cm^{-1}$. The comparison analysis is therefore limited to wavenumbers greater than the first Langley calibration point covered by the Menang-ESS, namely $\nu > 4200\ cm^{-1}$.
The exchange of the ESS has only minor influence on the continuum results as visible in Fig. 3 and Fig. 4 (which uses a linear scale to show the situation in the window regions). On average, the continuum absorption coefficient determined using the Menang-ESS (blue data points) differs from the results obtained with the Kurucz-ESS (black data points) by 11.1 % of the continuum uncertainty estimate. We use this difference as an estimate of the ESS-related continuum uncertainty and include it as an additional contribution in the continuum uncertainty budget presented in the companion paper Part I.

The following text was added to the manuscript to discuss the ESS-induced uncertainty of the continuum results (Page 7, line 12):
"As outlined in the companion paper Part II, recent studies on the NIR ESS have yielded results that feature differences of up to 5-10% (see e.g. Menang et al, 2013; Bolsee et al, 2014; Thuillier et al., 2014, 2015; Weber et al. 2015). Furthermore, the number of solar lines differs significantly e.g. between the ESS versions of Kurucz (2005) and Menang et al., (2013). To investigate the influence of inaccuracies in the ESS on the continuum results, the continuum retrieval was repeated using the ESS determined by Menang et al. (2013) instead of the ESS by Kurucz (2005) that was used to generate the results presented in Fig. 2. This is a good test to assess the sensitivity of the results to ESS uncertainty since these ESS versions differ by about 5 %, while recent ESS results generally feature differences of up to ±5 % compared to the ESS of Kurucz (2005). Note that the Menang et al. (2013) ESS only covers the spectral region $\nu > 4000\ cm^{-1}$. The comparison is therefore restricted to $4233\ cm^{-1} < \nu < 7800\ cm^{-1}$, which corresponds to the first Langley point covered by the Menang et al. (2013) ESS and the maximum wavenumber value covered by our analysis. For this region, the median of the absolute value of the difference between the Menang et al. (2013) and Kurucz (2005) continuum results corresponds to 11.1% of the continuum uncertainty estimate. Therefore, ESS uncertainty does not constitute a major accuracy limitation of our analysis, which his due to the fact that the same ESS is used for both synthetic spectra calculation and the radiometric calibration presented in the companion paper Part II. The ESS-related continuum uncertainty was estimated from the difference of the Menang et al. (2013) and Kurucz (2005) results and included in the uncertainty budget as described in Sect. 3.2 (see also Fig. 9 of the companion paper Part I). For the spectral region $\nu < 4233\ cm^{-1}$, where no direct comparison is available, the ESS-induced continuum uncertainty was assumed to correspond to 11.1% of the remaining overall uncertainty as suggested by the median value in the spectral range $\nu > 4233\ cm^{-1}$."

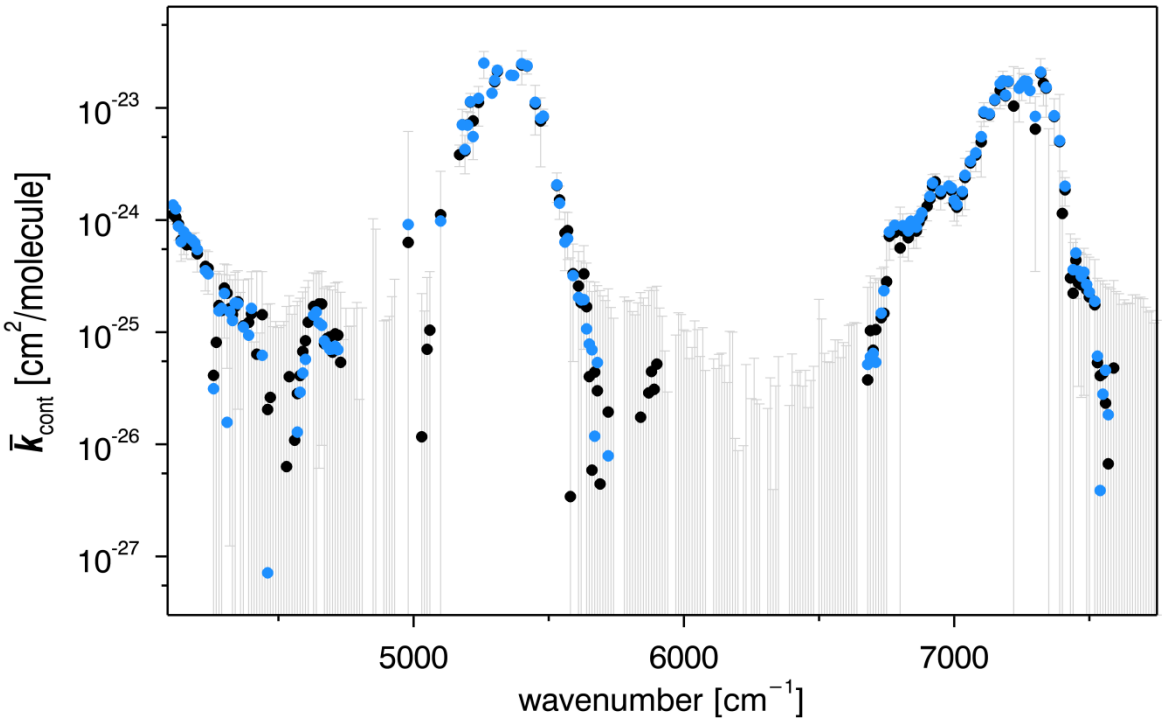

Fig. 3: Mean continuum absorption coefficient derived with the method and data set described in Sect. 3 using the ESS by Menang et al. (2013) (blue data points) and by Kurucz (2005) (black data points). The different ESS sources differ by about 5 % and many solar lines not present in Kurucz (2005) were included in Menang et al. (2013).

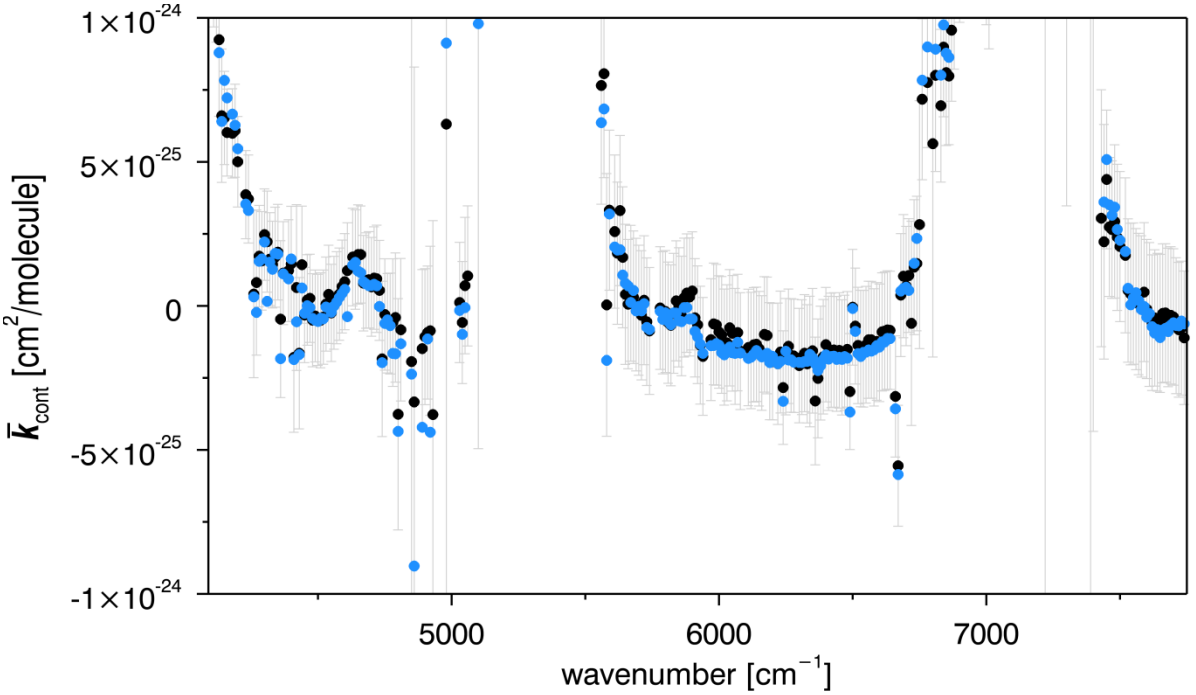

Fig. 4: Same as Fig. 3 but using a linear scale to show the results in the window regions.

2. The consistency between the residual method of deriving the optical depth could be compared with the slopes of the Langley plots in part II, as these are quasi - independent derivations of optical depth (and in particular, the Langley method does not require knowledge of $S_{actual)}$.

This consistency check had been performed by the authors but was omitted in the initial manuscript for the sake of brevity. The Langley results on the continuum are consistent with

the results derived from calibrated spectra throughout 98.0 % of the 2500 to 7800 cm^{-1} spectral range (see figure below) and the corresponding results are included in the revised manuscript as an appendix (Appendix C).

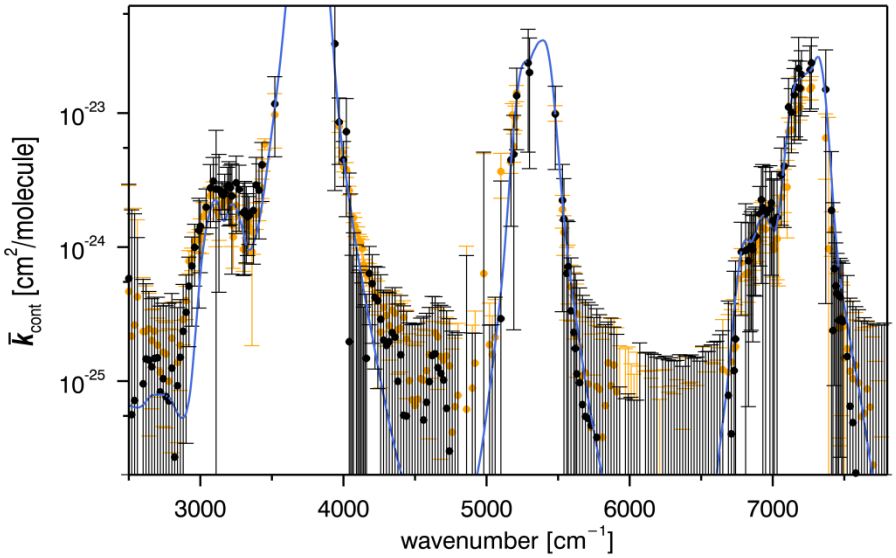

Fig. C1.: Mean continuum absorption coefficient $\bar{k}_{cont}$ determined from 12 December 2013 spectra using the Langley method and corresponding 2 σ uncertainties (black). Results are compared to the calibrated method for the same spectral dataset (orange) and the MT_CKD 2.5.2 model (blue).

3. We feel that the summary in the final two sentences of the abstract gives a somewhat misleading picture of the degree of agreement between the new observations and available laboratory measurements. For example, in Figure 1, it is difficult to see that the new measurements are in better agreement with the Bicknell measurements than the FTIR measurements of Ptashnik et al. (2012, 2013). From 5900 to 6600 cm^{-1}, the values derived in this paper, and listed in the Supplement, are almost universally negative, and therefore unphysical. In the 4700 cm^{-1} region, at the wavenumber of Bicknell's measurements (about 4670 cm^{-1}), the author's central estimate appears as close to the Ptashnik estimate as to Bicknell.

Even the comparison with the Mondelain et al. (2015) data is inconclusive. At wavenumbers just below 4250 cm^{-1}, where the authors' data have relatively small error bars, the data points tend to go in between the Mondelain et al. and Ptashnik et al. data. It is only at wavenumbers above 4250 cm^{-1} that the new data appear to fit better with Mondelain et al., but at these wavenumbers the new observations have too high uncertainties to allow firm conclusions; the upper errorbars nearly overlap the Ptashnik et al. data.

We feel that there would be greater clarity in the abstract if the situation near the band centre is separated from the situation in the window. In the band centres the disagreements between recent FTIR measurements (see especially Paynter et al. (2009)) and MT_CKD are known to be relatively small, compared to the situation in the windows; these near – band - centre regions constitute much of the "75%" that is referred to at 1(13). In the windows (e.g. 2800 – 3000 cm^{-1} and 4200 - 4500 cm^{-1}), it seems hard to sustain an argument that the new measurements are in any better agreement with MT_CKD than they are with the Ptashnik FTIR measurements.

The abstract was changed as follows to avoid a misleading impression on the degree of agreement (Page 1, line 15): "In the wings of water vapor absorption bands, our measurements indicate about 2-5 times stronger continuum absorption than MT_CKD, namely in the 2800 to 3000 cm^{-1} and 4100 to 4200 cm^{-1} spectral ranges. The measurements

are consistent with the laboratory measurements of Mondelain et al. (2015), which rely on cavity ring-down spectroscopy (CDRS), and the calorimetric-interferometric measurements of Bicknell et al. (2006). Compared to the recent FTIR laboratory studies of Ptashnik et al. (2012, 2013), our measurements are consistent within the estimated errors throughout most of the spectral range. However, in the wings of water vapor absorption bands our measurements indicate typically 2 – 3 times weaker continuum absorption under atmospheric conditions, namely in the 3200 to 3400 cm^{-1}, 4050 to 4200 cm^{-1}, and 6950 to 7050 cm^{-1} spectral regions."

Further comments
(coordinate system "page number(line number)")

1(18) and 8(3) We would say "typically a factor of 2 - 3 times higher". "5" seems an exaggeration to us.

Since differences by a factor of 5 between our results and the findings of Ptashnik et al. (2012, 2013) only occur at few spectral points, the manuscript was changed as suggested.

6(18) It would be useful to more clearly state how the solar absorption lines were defined. We assume these were based on the Kurucz ESS described in Part II. However, as noted by Menang et al. (2013) (using both an analysis of their own ground - based observations and using the ACE space - based measurements of Hase et al. (2010)), the Kurucz ESS does not include a number of solar lines that were detected in these two recent works.

The following more extensive discussion of solar line removal was included in the revised manuscript (Page 6, line 18):" ii) Regions around solar lines were excluded. This was implemented as an exclusion of all points for which the extra-atmospheric solar radiance according to the ESS of Kurucz (2005) is more than 0.5 % below the upper envelope. Note that recent studies indicate that many solar lines are missing in this ESS (see Menang et al., 2013). However, solar lines omitted in the ESS of Kurucz (2005) are discarded from further analysis by applying the selection criterion (i). As outlined in Sect. 4, a repetition of the continuum analysis using the ESS of Menang et al. (2013), which includes many additional solar lines only leads to very minor changes in the continuum results, thereby indicating that the solar line removal scheme according to criteria (i) and (ii) is appropriate."

7(1-8) We feel it would be useful to produce a plot that showed k_{cont} using both the linear+constant and the purely constant scaling. At present, the paper has only one figure, and so this could easily be accommodated.

The analysis on page 7, line 1-8 covers the scaling of k_{cont} with respect to IWV, which furthermore depends on wavenumber. Therefore, a complete representation of the data either requires a three-dimensional plot with axes v, IWV, and k_{cont} or two-dimensional plots with axes IWV and k_{cont} for each of the 532 wavenumber bins. The first alternative is not suitable for a figure due to the large number of data points in one plot, while the second alternative requires excessive space. We therefore decided to include supplementary figures in the revised manuscript (Fig. A3) showing the measured k_{cont} and the best fit linear and constant scaling for three representative wavenumber bins: 7200 cm^{-1} (within water vapor absorption band), 4100 cm^{-1} (wing of band), and 2700 cm^{-1} (window region).

7(29-37) We are unclear why two different temperature dependencies are employed, depending on which laboratory data is used, and what impact it has. Also we were unsure

why the MT-CKD temperature dependence was considered more appropriate for some sets than others. It would be useful to see the impact of using a common temperature dependence with all data sets, to establish how much effect this has on the results.

In the updated version of Fig. 2, the temperature dependence of Rädel et al. (2015) is used for all laboratory measurements to improve consistency.

7(40) It may be useful to plot the Paynter and Ramaswamy (2014) data as well as the Baranov and Lafferty (2011) observations.

The data of Paynter and Ramaswamy (2014) was added to Fig 2. The following discussion of the results of Paynter and Ramaswamy (2014) was added to the manuscript (Page, line):
"The comparison of our results to the BPS_MTCKD 2.0 continuum proposed by Paynter and Ramaswamy (2014) is mostly equivalent to the comparison to MT_CKD. This is due to the fact that the BPS_MTCD 2.0 foreign continuum, which constitutes the dominant contributor for the dry atmospheric conditions encountered in our data set, was mostly adopted from MT_CKD. Exceptions include the spectral regions from 2500 to 3000 cm^{-1}, 5200 to 5600 cm^{-1}, and 6800 to 7000 cm^{-1}, where our results show better consistency with the MT_CKD 2.5.2 model"
The intention of Fig. 2 (previously Fig. 1) was to present a comparison of our results to measurements which comprise a quantification of the self and foreign continuum at or below room temperature using the same or a very similar experimental setup. To clarify this intention, the following text was added to the manuscript (Page 6, line 30):
"The figure includes laboratory measurements carried out at or below room temperature which provided constraints on both the self and foreign continuum using the same or a very similar experimental setup."
We therefore did not include the results of Baranov (2011) and Baranov and Lafferty (2011) in Fig. 2 which were obtained at > 311 K for the self continuum and > 326 K for the foreign continuum. However, the following discussion of the results of Baranov (2011) and Baranov and Lafferty (2011) was added to the revised manuscript (Page 8, line 4): Further FTIR laboratory measurements were carried out by Baranov and Lafferty (2011) at temperatures of 311 - 363 K on the self continuum and by Baranov (2011) at 326 – 363 K on the foreign continuum at v < 3500 cm^{-1}. The results of these studies generally agree well within the estimated errors with the findings of Ptashnik et al. (2012, 2013).

8(7) There is misleading phrase. There were no "narrow line – like features in the continuum" reported e.g. by Ptashnik et al. (2011); those features were 60 cm^{-1} (FWHM) broad continuum peaks.

The misleading text was removed from the manuscript.

8(9-16) We largely agree with the statements here, but we believe it should be added that the assumption that the foreign continuum has no temperature dependence has not been tested at atmospheric temperatures in the laboratory. And it is that foreign continuum which dominates in the wings of water vapour absorption bands (in particular in the 3200 – 3400 and 4000 – 4200 cm-1 regions) where the large and more certain disagreement with FTIR - based results of Ptashnik et al. (2012) is noted by the authors.

The following text was added to the manuscript (Page 8, line 16):
"Note, however that the assumption that the foreign continuum has no significant temperature dependence, which was used in the data analysis, has not been robustly confirmed by measurements under atmospheric conditions yet. Due to the dominant role of the foreign continuum in the wings of water vapor absorption bands, inaccuracies in the foreign continuum temperature dependence would have a significant influence on the conversion of the findings of Ptashnik et al. (2012, 2013) to atmospheric temperatures."

8(31) We think that it should be pointed out that there are regions with rather good agreement with Ptashnik et al. (and better than with MT_CKD), particularly around 3000 cm^{-1}

The following text was added to the manuscript (Page 8, line 31): "There are also several regions where our results are in good agreement with the findings of Ptashnik et al. (2012, 2013), most notably around 3000 cm^{-1}."

13(1) It is rather hard to see the uncertainty bars, especially where they overlap with other data. Perhaps these could be drawn in a bolder format?
In addition, we suggest that an additional plot is needed to make clear to the reader that many of the derived values are negative/unphysical; this, of course, cannot be done in a plot with a logarithmic axis.

A darker color and bolder format was used for the error bars in the revised manuscript. The additional Fig. 4 now shows a non-logarithmic representation of the continuum results.

The Zugspitze radiative closure experiment for quantifying water vapor absorption over the terrestrial and solar infrared. Part III: Quantification of the mid- and near-infrared water vapor continuum in the 2500 to 7800 cm^{-1} spectral range under atmospheric conditions

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Andreas Reichert and Ralf Sussmann

Karlsruhe Institute of Technology, IMK-IFU, Garmisch-Partenkirchen, Germany

Correspondence to: A. Reichert (andreas.reichert2@kit.edu)

Abstract. We present a first quantification of the near-infrared (NIR) water vapor continuum absorption from an atmospheric radiative closure experiment carried out at Mt. Zugspitze (47.42° N, 10.98° E, 2964 m a.s.l.). Continuum quantification is achieved via radiative closure using radiometrically calibrated solar FTIR absorption spectra covering the 2500 to 7800 cm^{-1} spectral range. The dry atmospheric conditions at the Zugspitze site (IWV 1.4 to 3.3 mm) enable continuum quantification even within water vapor absorption bands, while upper limits for continuum absorption can be provided in the centers of window regions. Throughout 75 % of the 2500 to 7800 cm^{-1} spectral range, the Zugspitze results are agree within our estimated uncertainty with the widely used MT_CKD 2.5.2-model (Mlawer et al., 2012). ~~Notable exceptions ar~~In the wings of water vapor absorption bands, our measurements indicate about 2-5 times stronger continuum absorption than MT_CKD, namely in ~~e~~ the 2800 to 3000 cm^{-1} and 4100 to 4200 cm^{-1} spectral ranges~~, where our measurements indicate about 5 times stronger continuum absorption than MT_CKD~~. The measurements are consistent with the laboratory measurements of Mondelain et al. (2015), which rely on cavity ring-down spectroscopy (CDRS), and the calorimetric-interferometric measurements of Bicknell et al. (2006). Compared to the recent FTIR laboratory studies of Ptashnik et al. (2012~~,) and (~~2013), our measurements ~~indicate~~ are consistent within the estimated errors throughout most of the spectral range. However, in the wings of water vapor absorption bands our measurements indicate typically 2 – ~~5~~3 times weaker continuum absorption under atmospheric conditions~~ in the wings of water vapor absorption bands~~, namely in the 3200 to 3400 cm^{-1}, 4050 to 4200 cm^{-1}, and 6950 to 7050 cm^{-1} spectral regions.

1 Introduction

Atmospheric water vapor is the most important contributor to the absorption of incoming solar radiation in the near infrared (NIR, 4000—14000 cm^{-1}) (Kiehl and Trenberth, 1997). Water vapor absorption comprises both the effect of spectral line absorption and the broadband so-called continuum absorption (e.g. Shine et al., 2012). Depending on the atmospheric state and the choice of continuum model, up to 6% of the clear-sky water vapor absorption can be attributed to the continuum (Paynter and Ramaswamy, 2011). Consequently, ~~exact~~ quantitative knowledge of this contribution is a prerequisite for realistic atmospheric radiative transfer calculations employed e.g. in climate models (Paynter and Ramaswamy, 2014; Rädel et al., 2015; Turner et al., 2012).

However, the NIR atmospheric water vapor continuum currently still lacks sufficient experimental constraints. Recently, a number of laboratory studies based on different experimental techniques investigated this open question. Several efforts were made to quantify continuum absorption, including the contributions of both the self and foreign-broadened continuum~~both contributions to the continuum absorption, self and foreign continuum~~. Several studies made use of cell measurements with ~~FTIR (Fourier Transform Infrared) spectrometers~~ grating spectrometers (e.g. Burch 1982; 1985; Burch and Alt 1984~~) ;~~or FTIR (Fourier Transform Infrared) spectrometers (Baranov et al., 2008; Baranov and Lafferty, 2011; Paynter et al., 2009;

Ptashnik et al. 2011; 2012; 2013; and 2015). Furthermore, a number of spectral regions were covered by cavity ring-down spectroscopy (CRDS) measurements (Cormier et al. 2002, 2005; Mondelain et al., 2013, 2014~~;~~, 2015), by the related technique of optical feedback cavity enhanced spectroscopy (OF-CEAS, Ventrillard et al., 2015) and by calorimetric-interferometric measurements (Fulghum and Tilleman, 1991; Bicknell et al., 2006). However, no consensus has been reached among these studies. As noted e.g. by Mondelain et al. (2014) and Ptashnik et al. (2013), the individual results feature differences far beyond the respective uncertainty estimates whose attribution to causative processes remains tentative. A further challenge for laboratory studies is that they are ~~normally not feasible at representative atmospheric conditions since they~~ typically ~~rely e.g. on~~ carried out at higher temperatures than those encountered in the atmosphere in order to detect the weak continuum absorption in the limited optical path length of the cells. Note that CDRS and related techniques in principle enable measurements at atmospheric temperature (see e.g. Cormier et al., 2005) but such measurements are not yet available for many spectral regions). To date, the temperature dependence of the self-continuum has been investigated by measurements in a number of spectral regions (e.g. Cormier et al., 2005; Mondelain et al., 2014; Ptashnik et al., 2011; Ventrillard et al., 2015). However, the remaining uncertainty of the self-continuum temperature dependence (see e.g. Paynter and Ramaswamy, 2011) and the lack of measurements of the foreign continuum temperature dependence cause considerable uncertainties in the ~~still lacks observational constraints (see e.g. Paynter and Ramaswamy, 2011). This makes~~ application of the laboratory results on atmospheric radiative transfer calculations ~~non-straightforward~~.

The continuum has been investigated for atmospheric conditions using measurements of atmospheric emitted infrared radiance for other spectral regions (e.g. Tobin et al., 1999; Rowe and Walden, 2009). However, atmospheric measurements are available only for a fraction of the spectral region covered by this study (Newman et al. 2011; 2400 to 3200 cm-1), while for the remaining interval from 3200 to 7800 cm^{-1}, no atmospheric measurements have been reported. ~~A~~ A validation of continuum absorption strength under atmospheric conditions is therefore highly desirable to address these shortcomings. To this aim, we conducted a radiative closure experiment with the objective of quantifying the NIR water vapor continuum absorption from atmospheric measurements. The study is carried out at the high-altitude Zugspitze site and relies on the solar FTIR measurements implemented at this site (Sussmann and Schäfer, 1997) in the framework of the Network of the Detection of Atmospheric Composition Change (NDACC, www.ndacc.org). While such atmospheric closure studies enable to avoid some limitations of laboratory measurements as outlined above, they are also subject to a number of major challenges: absorption in the NIR due to aerosols can become comparable to the magnitude of the water vapor continuum absorption of interest (Ptashnik et al., 2015) and requires an accurate separation of continuum and aerosol contribution. Furthermore, the characterization of the atmospheric state (e.g. IWV, water vapor profile, temperature profile, and further trace gas column amounts) is more challenging and typically less accurate than the characterization of experimental conditions in a laboratory study.

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This paper is part of a three-paper series about different aspects of the Zugspitze radiative closure experiment. The first paper, ~~ther~~hereafter referred to as Part I (Sussmann et al., 2016, same issue), describes the instrumental setup, evaluates the sensitivity of the closure experiment in the far infrared (FIR, 2—667 cm^{-1}), the mid-infrared (MIR, 667—4000 cm^{-1}), and the NIR, and provides results on the FIR water vapor continuum. A novel radiometric calibration method for solar FTIR spectra in the NIR is presented in a second paper, referred to as Part II (Reichert et al., 2016, same issue). Part III (this paper) contains the NIR continuum quantification method and results. Continuum quantification in the NIR is achieved comparing calibrated radiance spectra, obtained with the method presented in Part II, to radiative transfer model calculations. The results derived from our data set are presented and compared to results from laboratory studies as well as the widely used MT_CKD 2.5.2 continuum model (Mlawer et al., 2012).

This paper is structured as follows: Section 2 contains an overview of the instrumental setup used in the closure experiment. Section 3 outlines the method for water vapor continuum quantification. In Sect. 4, the results obtained with this method are presented and compared to previous studies. Finally, Sect. 5 contains a summary and conclusions.

2 Setup of the closure experiment

The closure experiment relies on a quantitative comparison of measurements of spectral radiance with synthetic spectra calculated using the line-by-line radiative transfer model (LBLRTM, Clough et al., 2005). Spectral line parameters were set according to the aer_v3.2 line list provided alongside the LBLRTM model. Water vapor continuum absorption is then quantified via the spectral residuals, i.e. the difference between simulated and measured spectra. We adopt the definition of the water vapor continuum given in Turner et al. (2010), i.e. water vapor continuum is defined as all absorption by water vapor exceeding a Voight line shape within ± 25 cm^{-1} of each line center minus the value of the Voight line shape at ± 25 cm^{-1} ("plinth").

The instruments used in the Zugspitze radiative closure experiment ~~u~~and the related uncertainties are described in detail in Part I. In summary, spectral radiances in the NIR are measured using a solar FTIR spectrometer setup at the Zugspitze (47.42° N, 10.98° E, 2964 m a.s.l.) summit observatory (Sussmann and Schäfer, 1997). Radiative calibration of measured spectra is achieved via a novel calibration method presented in Part II, which relies on a combination of the Langley method and measurements of a medium-temperature blackbody source.

The atmospheric state at the time of the radiance measurements is required as an input to the LBLRTM radiative transfer calculations. To enable accurate quantification of the water vapor continuum from spectral residuals, the atmospheric state has to be constrained precisely using a number of additional measurements listed in the following.

Vertically integrated water vapor (IWV) constitutes the key input parameter and is derived directly from the solar FTIR spectra (e.g. Sussmann et al. 2009; Schneider et al., 2012). Temperature and pressure profiles are taken from four-times-daily National Center for Environmental Prediction (NCEP) resimulation data. NCEP resimulation data is also used to constrain the shape of the water vapor profile. Column-averaged mixing ratios of CO_2, CH_4, and N_2O are measured using the nearby Garmisch TCCON (Total Carbon Column Observing Network) solar FTIR instrument (Sussmann and Rettinger, 2014). O_3 columns were constrained by combined Brewer/Dobson measurements made at the nearby Hohenpeissenberg observatory (Köhler, 1995).

Aerosol optical depth (AOD) has to be constrained precisely in order to enable continuum quantification in the wings of the strong NIR water vapor bands and within window regions. A great advantage of the Zugspitze site is that AOD is typically very low, i.e. the AOD for the Zugspitze dataset is about a factor of 10 lower than at typical lowland mid-latitude sites. The AOD levels encountered in our closure data set (for data set description and selection criteria see Sect. 3.3) are in the range 0.0005 - 0.00075 at 2500 cm^{-1} and in the range 0.0024 - 0.0032 at 7800 cm^{-1} at airmass 1. AOD was measured using the SSARA-Z (Sun-Sky Automatic Radiometer - Zugspitze) sun photometer (Toledano et al., 2009) developed by the Meteorological Institute of the University of Munich and set up at Schneefernerhaus (2675 m a.s.l., 680 m horizontal distance to the Zugspitze solar FTIR). The instrument ~~disposes of~~ includes 13 spectral channels from 340 to 1640 nm. Only information from 5 channels ~~centered at~~ whose central wavelengths are in the spectral region between 439.6 ~~to~~and 781.1 nm was used in the analysis. The exact filter wavelengths and full width at half maximum (FWHM) values of these channels are listed in Table 1. The reason for the channel selection is that in the ultra-violet (UV) to visible range, water vapor continuum absorption can be considered negligible compared to AOD, whereas for the NIR channels continuum absorption will lead to biased AOD results. The channels below 440 nm were excluded since the high influence of Rayleigh scattering in the UV leads to increased AOD uncertainties.

The data analysis of the SSARA-Z measurements was implemented similar to the approach outlined by Toledano et al. (2009). In detail, we used standard Langley calibration for cloud-free periods. Rayleigh scattering was accounted for using the formula given by Bodhaine et al. (1999). In the analysis, a Gaussian shape was assumed for the filter transmissivity curves. The influence of absorption by O_3 was subtracted as outlined in Guyemard et al. (1995). NIR AOD was then deduced by assuming AOD wavelength dependence according to the Ångstrom relation:

$$\tau(\lambda) = b \cdot \lambda^{-\alpha}, \quad (1)$$

where τ designates AOD. The Ångstrom exponent α and scaling b are determined by a fit to the UV/visible AOD measurements. More sophisticated descriptions of the AOD wavelength dependence such as the relation given by Molineaux et al. (1998) may be used instead of Eq. (1). However, the number of sun photometer wavelength channels included in our analysis is not sufficient to place tight constraints on the higher number of parameters used in such models. Furthermore, the very low AOD at Zugspitze leads to high relative errors in the AOD determined from sun photometer measurements, which removes the benefits of more advanced models compared to Eq. (1).

~~The following contributions were included in the calculation of the AOD uncertainty:~~ The AOD uncertainty comprises several contributions: First of all, the AOD determined from the sun photometer measurements ~~are~~ is affected by uncertainty in the radiance measurements. This uncertainty contribution was set according to the 2-σ radiance measurement noise. The calibration uncertainty ensues from the uncertainty of the Langley fit. Additional uncertainty arises from the Rayleigh scattering correction, where central wavelength and FWHM errors of optical filters and atmospheric pressure errors contribute. The treatment of O_3 absorption is also prone to additional errors, due to filter parameter and O_3 column errors. In addition to these contributions, further uncertainty is induced by the fit to Eq. (1) that enables constraining the NIR AOD from the UV/visible measurements. The overall AOD uncertainty that ensues from these contributions for our data set at air mass 1 is <0.0015 at 2500 cm^{-1} and <0.0025 at 7800 cm^{-1}.

3 NIR continuum determination

3.1. Method overview

The aim of this study is to constrain the NIR water vapor continuum absorption under atmospheric conditions. We make use of the radiative closure experiment setup at the Zugspitze observatory that is described in detail in Part I. Generally, radiative closure experiments comprise a quantitative comparison of spectral radiance measurements to synthetic spectra. The strategy for water vapor continuum quantification employed in this study relies on radiometrically calibrated solar FTIR spectra in the 2500 to 7800 cm^{-1}-range. An alternative method that has been proposed by Mlawer et al. (2014) and relies on the Langley method is presented in Appendix C.

Spectra were recorded with the solar FTIR instrument described in Sect. 2 and Part I, using no optical filter, a spectral resolution of 0.02 cm^{-1} (resolution is defined as 0.9/optical path difference), and averaging over 4-8 scans which leads to a 75-150 s repeat cycle per spectrum. The measured spectra are radiometrically calibrated by means of the calibration method outlined in Part II. Briefly, the calibration approach relies on Langley calibration in suitable spectral windows with little atmospheric absorption. In addition to the Langley technique that enables highly accurate calibration in selected windows, the shape of the calibration curve between the windows is constrained using spectral radiance measurements of a high-temperature blackbody source. The calibration uncertainty achieved with this novel method is 1-1.7 % (2 σ) throughout the spectral range considered. Synthetic radiance spectra are generated using the LBLRTM radiative transfer model. Figure 1 shows the mean measured and synthetic radiance spectra for the closure data set that will be presented in Sect. 3.3. The atmospheric state used as an input to the calculations was set based on the measurements described in Sect. 2. Given the calibrated spectral radiance measurements and the synthetic spectra, radiance residuals $\Delta \boldsymbol{I}$ can then be calculated for a set of spectra selected according to the criteria that will be presented in Sect. 3.3

$$\Delta \boldsymbol{I} = \boldsymbol{I}_{\mathrm{FTIR}} - \boldsymbol{I}_{\mathrm{LBLRTM,\ no\ continuum}} \cdot e^{-\mathrm{AOD}}, \quad (2)$$

where $\boldsymbol{I}_{\mathrm{FTIR}}$ designates the radiometrically calibrated solar FTIR spectra, $\boldsymbol{I}_{\mathrm{LBLRTM,\ no\ continuum}}$ the synthetic LBLRTM spectra not including continuum absorption and AOD the aerosol optical depth. Continuum optical depth $\boldsymbol{\tau}_{\mathrm{cont}}$ is calculated from the spectral residuals as follows:

$$\boldsymbol{\tau}_{\text{cont}} = -\ln\left(\frac{\Delta \boldsymbol{I}}{\boldsymbol{I}_{\text{LBLRTM, no continuum}} \cdot e^{-\text{AOD}}} + 1\right). \quad (3)$$

After calculation of the continuum optical depth (OD), absorption coefficients were derived from these results. The continuum OD $\boldsymbol{\tau}_{\text{cont}}$ is linked to continuum absorption coefficient $\boldsymbol{k}_{\text{cont}}$ as follows

$$\boldsymbol{\tau}_{\text{cont}} = m \cdot \int_{h_{\text{obs}}}^{\infty} \boldsymbol{k}_{\text{cont}}(T, n_{\text{wv}}, n_{\text{air}}) \cdot n_{\text{wv}} dh, \quad (4)$$

where m designates the relative air mass, h_{obs} the altitude of the observing instrument, n_{wv} the water vapor number density, and n_{air} the dry air number density.

$\boldsymbol{k}_{\text{cont}}$ can be further decomposed in self- and foreign continuum contributions according to

$$\boldsymbol{k}_{\text{cont}} = \boldsymbol{c}_{\text{s}} \cdot \frac{\rho_{\text{H}_2\text{O}}}{\rho_0} + \boldsymbol{c}_{\text{f}} \cdot \frac{\rho_{\text{air}}}{\rho_0}, \quad (5)$$

where $\boldsymbol{c}_{\text{s}}$ and $\boldsymbol{c}_{\text{f}}$ designate the self- and foreign continuum coefficients and $\rho_{\text{H}_2\text{O}}$, ρ_{air}, and ρ_0 are the densities of water vapor, dry air and a reference density, respectively. In detail, $\rho_0 = P_0/(k_{\text{b}} T_0)$, where P_0 = 1013 mbar, k_{b} is the Boltzmann constant, and T_0 = 296 K. In addition to their different dependence on water vapor density according to Eq. 5, self- and foreign broadened continua are characterized by their distinct temperature dependence: while the self continuum shows strong negative temperature dependence, the foreign continuum is assumed to have no or only weak temperature dependence.

The separation of $\boldsymbol{k}_{\text{cont}}$ in self- and foreign continuum contributions from atmospheric measurements is challenging. In principle, an assignment to self- and foreign continuum is possible using a large set of measurements covering a wide range of atmospheric conditions, i.e. IWV and temperature. However, the available data does not permit such an assignment given the sensitivity of our setup as discussed in Sect. 4. Therefore, in the following, we characterize continuum strength using the mean continuum absorption coefficient $\overline{\boldsymbol{k}}_{\text{cont}}$, defined as follows:

$$\overline{\boldsymbol{k}}_{\text{cont}} = \frac{\int_{h_{\text{obs}}}^{\infty} \boldsymbol{k}_{\text{cont}}(T, n_{\text{wv}}, n_{\text{air}}) \cdot n_{\text{wv}} dh}{\int_{h_{\text{obs}}}^{\infty} n_{\text{wv}} dh,} = \frac{\boldsymbol{\tau}_{\text{cont}}}{m \cdot \text{IWV}} \quad (6)$$

Low-uncertainty constraints on $\overline{\boldsymbol{k}}_{\text{cont}}$ can only be placed in a number of spectral windows. The selection of such suitable windows is outlined in Sect. 3.4. The continuum results for each spectrum were computed as the median of $\overline{\boldsymbol{k}}_{\text{cont}}$ in all selected spectral windows within 10 cm^{-1}-wide bins. Finally, an error-weighted mean of $\overline{\boldsymbol{k}}_{\text{cont}}$was calculated from the set of 52 spectra selected according to the criteria listed in Sect. 3.3. The uncertainty estimate of the continuum results is presented in Sect. 3.2.

3.2. Uncertainty estimate

An interpretation of the residual OD and assignment to causative absorption processes requires a comprehensive uncertainty budget of the closure experiment. The uncertainty estimate of our experimental setup is described in detail in Part I except for contributions only relevant for the NIR closure measurements. The total residual uncertainty and its various contributions are also shown in Part I, Fig. 5. A description of the NIR-specific contributions and a brief outline of the remaining sources of uncertainty are given below. All uncertainty values are quoted on 2-σ confidence level.

i) Absorption line parameter uncertainties of water vapor and other absorbing species. These uncertainties were set to the mean value of the uncertainty range specified by the error codes provided in the line parameter file (aer_v3.2) provided alongside the LBLRTM model. Line parameter uncertainties are the dominant contribution to the error budget within absorption bands.

ii) A further significant contribution to the error budget results from the IWV measurement uncertainty. The IWV precision was set to 0.8 %, the bias to 1.1 % according Schneider et al. (2012). The uncertainty resulting from NCEP water vapor profile shape errors was estimated using a comparison of NCEP profiles to radiosonde data (see Part I for details).

iii) The OD uncertainty resulting from NCEP temperature profile errors was deduced from a temperature error covariance matrix estimate for the NCEP resimulation profiles. The error covariance matrix estimate was constructed from the comparison of coincident NCEP profiles to a radiosonde campaign conducted at the site (see Part I for details).

iv) Column uncertainties for further trace gases (e.g. CO_2, CH_4, N_2O and O3) are also included in the uncertainty estimate. The respective column accuracies are listed in Part I (Tab. 2 therein).

v) The AOD uncertainty is of crucial importance for the OD uncertainty budget in the window regions. As outlined in Sect. 2, the AOD uncertainty at air mass 1 is < 0.0025 for the closure data set throughout the 2500 to 7800 cm^{-1}-range.

The uncertainty contributions i) to v) listed above are linked to the accuracy of the atmospheric state input for LBLRTM calculations. Aside from that, an additional group of error contributions stems from the solar FTIR spectral radiance measurements:

vi) The radiance uncertainty due to the radiometric calibration is about 1 – 1.7% and is described in detail in Part II.

vii) A further uncertainty contribution results from the solar FTIR measurement noise. It is determined directly from solar FTIR spectra and is among the few uncertainty contributions in the closure experiment of strictly statistical character. It is therefore largely reduced by taking mean results from a larger set of spectra.

viii) Ice layer formation on the liquid nitrogen cooled InSb detector can occur in case of leaks in the detector's vacuum enclosure. Ice formation leads to additional absorption in certain spectral regions, most notably in the 3000 to 3400 cm^{-1}-range. The uncertainty contribution by varying ice absorption was estimated using lamp spectra routinely recorded with the solar FTIR. Variations in ice absorption during the time period covered by the experiment can be detected as a change of the ratio of measured signal outside and inside the ice absorption band. The maximum variation of this ratio detected in the lamp spectra (1.6 %) was taken as an estimate of the error due to ice absorption.

ix) Only a fraction of the solar tracker mirrors is covered by the instrument's field of view (FOV). Due to non-ideal alignment of optical elements, the exact location of the area observed by the instrument on the mirror changes depending on the azimuth and elevation of the instrument's line of sight. The reflectivity of the tracker mirrors features spatial inhomogeneity due to dirt and aging effects. In combination with the moving area covered by the FOV, this results in a variation in measured radiance which leads to spurious variations in the measured OD. An estimate of this uncertainty contribution can be gained using an outgoing laser beam aligned with the instrument's optical axis that enables constraining the mirror area covered by the FOV depending on the instrument's azimuth and elevation. A detailed description of this analysis is given in Part II, Sect. 4.1.

x) The uncertainty due to inaccuracies in the ESS was estimated from repeating the continuum retrieval using the ESS versions by Kurucz (2005) and Menang et al. (2013), which differ by about 5 % (see Sect. 4). The corresponding uncertainty contribution corresponds to 11.1% of the remaining continuum uncertainty budget on average (see also Fig. 9 of the companion publication Part I).

3.3. Spectra selection

We analyzed spectra recorded under cloud-free conditions in the December 2013 – February 2014 period. Due to inaccuracies in the air mass calculation at high solar zenith angle, air mass was required to be below $m = 9.0$.

In Sect. 3.2, we outlined a source of radiance error in the solar FTIR measurements due to the pointing variation on the tracker mirrors and give an estimate of this contribution. For spectra included in the closure data set, this uncertainty contribution was required~~uested~~ to be negligible compared to other sources of uncertainty, in detail the selection threshold was set to a maximum radiance error of 0.1%. These selection criteria lead to a final dataset of 52 selected solar FTIR spectra covering an IWV range from 1.4 to 3.3 mm for which the continuum results are presented in Sect. 4. The mean atmospheric state of the closure data set is listed in Appendix A.

3.4. Micro-window selection

To select suitable windows for continuum quantification, a number of selection criteria were applied to the spectra. Several criteria make use of upper or lower envelopes to the spectra, which were constructed as follows: The upper/lower envelope is defined as the linear interpolation between the highest/lowest values encountered within each 10 cm^{-1}-wide wavenumber bin. In detail, the following filtering criteria were applied to the spectra:

i) To avoid spectral regions affected by line absorption, only the spectral points with the lowest OD compared to the surrounding spectral region were used. In detail, only points for which the OD exceeds the lower envelope by less than the 2-σ OD uncertainty were used.

ii) Regions around solar lines were excluded. This was implemented as an exclusion of all points for which the extra-atmospheric solar radiance according to the ESS of Kurucz (2005) is more than 0.5 % below the upper envelope. Note that recent studies indicate that many solar lines are missing in this ESS (see Menang et al., 2013). However, solar lines omitted in the ESS of Kurucz (2005) are discarded from further analysis by applying the selection criterion (i). As outlined in Sect. 4, a repetition of the continuum analysis using the ESS of Menang et al. (2013), which includes many additional solar lines, only leads to very minor changes in the continuum results, thereby indicating that the solar line removal scheme according to criteria (i) and (ii) is appropriate.

iii) Only regions with low OD uncertainty are included. Therefore, we select points less than 10 % above the lower envelope to the uncertainty.

iv) In order to avoid biases of the retrieved continuum due to measurement noise, only regions with a signal-to-rms-noise ratio $s/n > 5$ were included.

The selection thresholds cited above were adjusted in order to provide sufficiently dense coverage with selected points while maintaining optimum selection quality. Different experimental setups may therefore require different selection threshold values. The final continuum OD results were computed as the median value of all selected spectral points within 10 cm^{-1}-wide bins.

4 Results

Figure 1 shows the mean continuum absorption coefficient $\overline{\boldsymbol{k}}_{\mathrm{cont}}$ determined from the Zugspitze dataset in comparison to the MT_CKD 2.5.2 model predictions and several recent laboratory studies. The figure includes laboratory measurements carried out at or below room temperature which provided constraints on both the self and foreign continuum using the same or a very similar experimental setup. The mean atmospheric state of the closure dataset is listed in Appendix A, while a table with our results for $\overline{\boldsymbol{k}}_{\mathrm{cont}}$ and the relative scaling of our results vs. the MT_CKD 2.5.2 predictions and associated uncertainties are available as supplementary material (Supplement A). The results shown in Fig. 2 are in very good agreement with the $\overline{\boldsymbol{k}}_{\mathrm{cont}}$-values derived using the Langley method as outlined in Appendix C. The assignment of the residual OD to water vapor continuum absorption was made based on two arguments: As outlined in Part I, great care was taken to construct a comprehensive uncertainty budget including thorough estimates of all relevant error contributions to the closure experiment. Therefore, contributions to the residual OD from other processes than water vapor continuum absorption far beyond the indicated error bars seem unlikely. Furthermore, the IWV dependence of the measured residual OD is consistent with that expected from water vapor continuum absorption. As $\overline{\boldsymbol{k}}_{\mathrm{cont}}$ includes contributions due to both foreign- and self continuum, it is expected to scale as the sum of a constant and a linear term with respect to water vapor density and therefore also with respect to IWV. The closure dataset covers an IWV range of 1.4 mm < IWV < 3.5 mm, which enables investigation of the IWV dependence of $\overline{\boldsymbol{k}}_{\mathrm{cont}}$. Due to the narrow range of atmospheric temperatures covered in the data set, temperature dependence of the self continuum can be neglected in this analysis. A fraction of 98.6 % of all measured continuum

absorption coefficients in the Zugspitze data set are consistent with a combination of constant and linear scaling with respect to IWV, i.e. with being caused by a combination of foreign- and self water vapor continuum. However, 94.2 % of the data are also consistent with a purely constant scaling, i.e. with being solely due to foreign continuum absorption.– This is due to the fact that at the atmospheric conditions covered by the data, in all spectral regions where continuum absorption is detectable beyond the experiment's sensitivity, the foreign continuum constitutes by far the dominant contribution, assuming that the partitioning in self- and foreign continuum given by the MT_CKD model is approximately correct. Note that this assumption has to be considered tentative since for both self- and foreign continuum the results of recent laboratory studies deviate from the MT_CKD model especially in window regions (e.g. Ptashnik et al, 2012, 2013; Mondelain et al., 2015). Examples of the measured $\bar{\boldsymbol{k}}_{\mathrm{cont}}$ and the best fit constant and linear scaling for wavenumber bins within water vapor bands, in the wings of bands and in window regions are shown in Fig. A3.

This analysis shows that the contribution of the self continuum is not unambiguously detectable due to the limited sensitivity of our experiment. We therefore provide values of the mean continuum absorption coefficient $\bar{\boldsymbol{k}}_{\mathrm{cont}}$ as defined by Eq. (6), including contributions from both self- and foreign continuum instead of the more commonly used continuum coefficients $\boldsymbol{c}_{\mathrm{s}}$ and $\boldsymbol{c}_{\mathrm{f}}$. If values for $\boldsymbol{c}_{\mathrm{f}}$ are required, further assumptions on the self continuum have to be made before subtracting this contribution. As an example, Supplement B to this manuscript contains a list of $\boldsymbol{c}_{\mathrm{f}}$-values for all spectral bins where $\boldsymbol{c}_{\mathrm{f}}$ exceeds the uncertainty estimate. The results were calculated from our measurements assuming the self continuum to be consistent with the MT_CKD model. Recent laboratory measurements (e.g. Ptashnik et al., 2013) suggest that this assumption may not be appropriate. However, alternative sources of the self continuum neither constitute a more robust estimate, given the inconsistencies between different laboratory results, the uncertainty of the self continuum temperature dependence and the fact that the foreign continuum is likely to be the dominant contribution to the overall continuum absorption for the dry atmospheric conditions of our study and the spectral windows covered by the measurements (see Fig. 1). –A 50 % uncertainty was assumed for the self continuum as suggested by Paynter et al. (2011) and is included in the uncertainty of $\boldsymbol{c}_{\mathrm{f}}$ in addition to the uncertainty budget presented in Sect. 3.2.

As outlined in the companion paper Part II, recent studies on the NIR ESS have yielded results that feature differences of up to 5-10% (see e.g. Menang et al, 2013; Bolsee et al, 2014; Thuillier et al., 2014, 2015; Weber et al. 2015). Furthermore, the number of solar lines differs significantly e.g. between the ESS versions of Kurucz (2005) and Menang et al., (2013). To investigate the influence of inaccuracies in the ESS on the continuum results, the continuum retrieval was repeated using the ESS determined by Menang et al. (2013) instead of the ESS by Kurucz (2005) that was used to generate the results presented in Fig. 2. This is a good test to assess the sensitivity of the results to ESS uncertainty since these ESS versions differ by about 5 %, while recent ESS results generally feature differences of up to ±5 % compared to the ESS of Kurucz (2005). Note that the Menang et al. (2013) ESS only covers the spectral region > 4000 cm^{-1}. The comparison is therefore restricted to 4233 cm^{-1} < ν < 7800 cm^{-1}, which corresponds to the first Langley point covered by the Menang et al. (2013) ESS and the maximum wavenumber value covered by our analysis. For this region, the median of the absolute value of the difference between the Menang et al. (2013) and Kurucz (2005) continuum results corresponds to 11% of the continuum uncertainty estimate. Therefore, ESS uncertainty does not constitute a major accuracy limitation of our analysis, which his due to the fact that the same ESS is used for both synthetic spectra calculation and the radiometric calibration presented in the companion paper Part II. The ESS-related continuum uncertainty was estimated from the difference of the Menang et al. (2013) and Kurucz (2005) results and included in the uncertainty budget as described in Sect. 3.2 (see also Fig. 9 of the companion paper Part I). For the spectral region ν < 4233 cm^{-1}, where no direct comparison is available, the ESS-induced continuum uncertainty was assumed to correspond to 11% of the remaining overall uncertainty as suggested by the median value in the spectral range ν > 4233 cm^{-1}.

The prediction of the MT_CKD 2.5.2 model is shown alongside our results for $\bar{\boldsymbol{k}}_{\mathrm{cont}}$ in Fig. 1. The MT_CKD 2.5.2-values of $\bar{\boldsymbol{k}}_{\mathrm{cont}}$ were computed in an analogous way as the values derived from our dataset, i.e. $\bar{\boldsymbol{k}}_{\mathrm{cont}}$ was calculated according to Eq. (6)

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for the set of atmospheric states encountered in the data set. The results shown in Fig. 1 represent the mean of the MT_CKD predictions for the set of selected measurements. Overall, there is good agreement of our results with the MT_CKD values. Consistency within a 2-σ range is observed for 75 % of the spectral range covered by our measurements. The most apparent discrepancy between MT_CKD and our results occurs in the 2800 to 3000 cm^{-1}-range, where our results are about a factor of 5 higher than the MT_CKD predictions. However, care has to be taken in the interpretation of this discrepancy since the 2800 to 3000 cm^{-1} spectral range coincides with a methane absorption band. Therefore, the accuracy of the continuum result in this range depends on whether the HITRAN error estimate for methane line parameters is correct and whether line coupling effects where treated in a sufficiently realistic way in the LBLRTM model. Further significant discrepancies ensue in the 4100 to 4200 cm^{-1} wavenumber region. The higher measurement results from the Zugspitze data indicate that the MT_CKD-model underestimates the continuum absorption in the wings of the 4000 to 5000 cm^{-1} window region. In the centers of water vapor absorption bands (i.e. ~5200-5400 cm^{-1} and ~7100-7300 cm^{-1}), our results are significantly lower than the MT_CKD-predictions for a number of spectral points. However, the continuum results in these regions are highly sensitive to accurate input and uncertainty estimates for IWV and water vapor line parameters. Therefore, the slight differences found in the band centers do not provide robust evidence for necessary adjustments of the MT_CKD model.

Figure 1 also includes a comparison of our results to several current laboratory studies using different experimental approaches for continuum quantification. For the comparison, $\bar{k}_{\mathrm{cont}}$-values were calculated for the same set of atmospheric states as our results using the continuum coefficients given in the respective studies. For the Mondelain et al. (2015) and Bicknell et al. (2006) results, we used the MT_CKD temperature dependence. Since the results of Bicknell et al. (2006) do not allow a dissociation of self from foreign continuum is not possible, we assumed the self-to-foreign ration suggested by the MT_CKD model to calculate the corresponding value of $\bar{k}_{\mathrm{cont}}$. ~~For the Ptashnik et al. (2012) and (2013) results~~The self continuum temperature dependence proposed by Rädel et al. (2015), which ~~, we employ the more sophisticated alternative temperature dependence proposed by Rädel et al. (2015)~~, which was deduced from the measurements of Ptashnik et al. (2011), was used for all laboratory studies. Note, however, that the importance of the continuum temperature dependence is limited (5 to 20 %, see below) for our dataset. This is due to the fact that no temperature dependence is assumed for the foreign continuum, which is by far dominant for most spectral regions given the dry atmospheric conditions encountered in our data set.

A fraction of the spectral range covered by this study, namely 2500-3200 cm^{-1}, was also included in the airborne measurements by Newman et al. (2011). Newman et al. (2011) conclude that the increase of the self continuum in MT_CKD 2.5 compared to MT_CKD 2.4 lead to reduced spectral residuals, while no firm conclusion can be drawn in the 2500 -3200 cm^{-1} –range on whether MT_CKD 2.5 or the results of Ptashnik et al. (2011) represent a more appropriate quantitative description of the water vapor self continuum. These findings are in agreement to the results of this study, given that both are not consistent with continuum absorption being weaker than indicated by MT_CKD 2.5. Our results show very good agreement with the CDRS-based measurements of Mondelain et al. (2015). Due to the dominant role of the foreign continuum in the 4100 to 4200 cm^{-1} spectral range, this agreement mainly corresponds to a comparison of the foreign continuum results of Mondelain et al. (2015) and our measurements. Therefore, our results are consistent with the finding of Mondelain et al. (2015) and Ptashnik et al. (2012) that the foreign continuum is underestimated by the MT_CKD model in this spectral region. For the spectral range examined by Bicknell et al. (2006) with calorimetric-interferometric measurements, only the upper limit of the continuum absorption is constrained by our data, which is consistent with all laboratory studies cited here. ~~In the window regions, our results show good consistency with the BPS-MTCKD 2.0 continuum proposed by Paynter and Ramaswamy (2014)~~The comparison of our results to the BPS_MTCKD 2.0 continuum proposed by Paynter and Ramaswamy (2014) is mostly equivalent to the comparison to MT_CKD. This is due to the fact that the BPS_MTCD 2.0 foreign continuum, which constitutes the dominant contributor for the dry atmospheric conditions encountered in our data set, was mostly adopted from MT_CKD. Exceptions include the spectral regions from

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2500 to 3000 cm^{-1}, 5200 to 5600 cm^{-1}, and 6800 to 7000 cm^{-1}, where our results show better consistency with the MT_CKD 2.5.2 model. The FTIR-based results of Ptashnik et al. (2012~~) and (~~, 2013) in combination with the temperature dependence proposed by Rädel et al. (2015) lead to higher absorption coefficients than our data in several spectral regions. Significant inconsistencies beyond the uncertainty range occur mostly in the wings of water vapor absorption bands, e.g. in the 3200 to 3400 cm^{-1}, and 4000 to 4200 cm^{-1} ranges as visible in Fig. 1. In these ranges the absorption coefficients provided by the FTIR laboratory measurements are typically a factor of 2-~~3~~5 higher compared to our data. Further FTIR laboratory measurements were carried out by Baranov and Lafferty (2011) at temperatures of 311 to 363 K on the self continuum and by Baranov (2011) at 326 to 363 K on the foreign continuum at $\nu < 3500$ cm^{-1}. The results of these studies generally agree well within the estimated errors with the findings of Ptashnik et al. (2012, 2013).

As noted e.g. by Ptashnik et al. (2015), weak lines not included in the line list used for the synthetic spectra calculation may bias the retrieved continuum results. This effect is largely reduced in our analysis due to the spectral selection criteria applied, namely the selection of low-OD windows as outlined in Sect. 3.4, criterion i). ~~A drawback of this selection scheme is that our analysis is not well suited for the detection of narrow line-like features in the continuum as reported e.g. by Ptashnik et al. (2011).~~

An issue not accounted for in our analysis is the uncertainty of the continuum temperature dependence, since an uncertainty estimate is provided neither for the MT_CKD nor the Rädel et al. (2015) relations. However, under the atmospheric conditions covered by our data set and assuming the MT_CKD self-to-foreign ratio, the self continuum contributes only 10 to 30 % to the total continuum absorption at the spectral points for which we detect significant continuum absorption. While no temperature dependence is assumed for the dominant foreign contribution, the temperature dependence of the self continuum changes the mean continuum absorption coefficient by 5 to 20 % within the spectral range considered here and assuming the Rädel et al. (2015) relation. Therefore, it seems unlikely that the differences between the results of Ptashnik et al. (2012) and (2013) and our data are solely due to inaccuracies in the continuum temperature dependence. Note, however that the assumption that the foreign continuum has no significant temperature dependence, which was used in the data analysis, has not been robustly confirmed by measurements under atmospheric conditions yet. Due to the dominant role of the foreign continuum in the wings of water vapor absorption bands, inaccuracies in the foreign continuum temperature dependence would have a significant influence on the conversion of the findings of Ptashnik et al. (2012, 2013) to atmospheric temperatures.

5 Summary and conclusions

We present~~ed~~ a quantification of the water vapor continuum absorption in the NIR spectral range (2500 to 7800 cm^{-1}) from an atmospheric radiative closure experiment. To our knowledge, prior to this study no precise constraints on the continuum absorption under atmospheric conditions were available ~~in~~for most of this spectral range. The mean continuum absorption coefficient was determined from a set of 52 solar FTIR spectra. The method to achieve continuum quantification relies on the use of radiometrically calibrated spectra obtained by the method presented in Part II. Continuum constraints are presented in the wings and some spectral windows in the centers of water vapor absorption bands. Due to the low IWV encountered throughout our measurement period, only the upper boundary of the continuum can be constrained in the centers of atmospheric windows.

The results show good consistency with the widely used MT_CKD 2.5.2 model, although they indicate a need for increasing the absorption strength compared to the model in some spectral regions such as the wings of water vapor absorption bands. Our results were compared to a number of recent laboratory studies using different experimental techniques. A first group of studies relies on FTIR cell measurements. Our data generally indicate lower continuum absorption than implicated by the studies of Ptashnik et al. (2012, ~~) and (~~2013) in combination with the self continuum temperature dependence given in Rädel

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at al. (2015). However, significant deviations from these studies only occur in the wings of water vapor absorption bands. There are also several regions where our results are in good agreement with the findings of Ptashnik et al. (2012, 2013), most notably around 3000 cm^{-1}. Further experimental techniques used for continuum quantification in laboratory experiments include CRDS. A comparison to the CDRS results of Mondelain et al. (2015) in the spectral region around 4250 cm^{-1} shows very good agreement to our findings. Bicknell et al. (2006) quantified continuum absorption using a calorimetric-spectrometric technique. While our results agree to the findings of Bicknell et al. (2006), their measurements cover spectral regions where only an upper limit for the continuum absorption can be deduced from our data.

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An assignment of the detected continuum absorption to self- and foreign continuum requires improvements of the experimental sensitivity or a data set covering a broader range of IWV values. The same is true for a detection of the continuum beyond the uncertainty limit in window regions which requires improved sensitivity or a data set covering higher IWV values. Aside from these limitations, our results provide a valuable foundation for an improved quantification of the NIR water vapor continuum under atmospheric conditions. Most notably, our analysis provides a tool for atmospheric validation of the predictions of current laboratory studies and the MT_CKD continuum model in the NIR spectral range. This is of crucial importance since the results of recent studies carried out using different experimental techniques show inconsistent results and to date no experimental validation under atmospheric conditions was available.

Appendix A: Mean atmospheric state of the closure data set

Appendix B: Supplementary figures

Appendix C: Continuum quantification using the Langley method

Langley measurements (see e.g. Liou, 2002 and Part II) are part of the radiometric the calibration method employed in this study which is presented in the companion paper Part II. These Langley measurements can be used to directly quantify the water vapor continuum as outlined by Mlawer et al. (2014), thereby offering a validation of the results obtained with the method presented in Sect. 3. For continuum quantification according to this strategy, spectrally resolved atmospheric optical depth is determined using the Langley method for all wavenumber values according to the analysis scheme presented in Part II. The atmospheric optical depth without water vapor continuum absorption is then calculated using the LBLRTM model using the atmospheric state input set as outlined in Sect. 3 and the companion paper Part I. Residual optical depth between Langley result and model calculation in spectral windows selected according to Sect. 3.4 is interpreted as water vapor continuum absorption.

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The OD uncertainty budget required for the window selection in the case of the Langley method is largely similar to the uncertainty estimate described in Sect. 3.2. Differences include the absence of the calibration uncertainty, which is replaced by the OD uncertainty of the Langley fit, including the uncertainty contribution due to air mass inaccuracies. Furthermore, the error due to changing ice absorption on the detector is not included in the error budget, since, by construction, the Langley method is only sensitive to atmospheric absorption. The mean continuum absorption coefficient is then calculated from the residual OD as described in Sect. 3.1.

Figure C1 shows the mean continuum absorption coefficients derived from the Langley measurements made on 12 December 2013 (black data points). The results are compared to the MT_CKD 2.5.2 model (blue line) and the results obtained with the calibrated method according to Sect. 3. (orange data points), which were calculated for the same set of 16 spectra used in the Langley method. Throughout 98.0 % of spectral range for which results from both methods are available, the absorption coefficients are consistent within the 2 σ error estimate. While in window regions both methods are equally suitable for

continuum quantification, larege errors due to air mass uncertainties make the use of the calibrated method presented in Sect. 3 preferable in the vicinity of water vapor absorption bands.

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Tables

Tab. 1: Central wavelength and FWHM of the sun photometer (SSARA-Z) filters used for AOD analysis.

λ [nm]	FWHM [nm]
439.6	9.7
498.7	12.3
531.9	11.2
672.5	10.9
781.1	9.7

Tab. A1: Mean atmospheric state of the closure data set~~.~~: pressure-, temperature- and water vapor density profiles. The data set was selected from Zugspitze solar FTIR spectra measured from Dec 2013 – Feb 2014 and contains 52 spectra. Spectra selection criteria are listed in Sect. 3.3.

altitude [km a.s.l]	P [mbar]	T [K]	$\rho_{water\ vapor}$ [g/m^3]	~~further parameters~~
2.964	714.074	270.570	1.270	~~IWV 2.26 mm~~
2.975	713.085	270.522	1.266	~~XCO_2 395.3 ppm~~
2.987	712.008	270.469	1.263	~~XCH_4 1781 ppb~~
3.009	710.036	270.372	1.256	~~XN_2O 311.8 ppb~~
3.032	707.982	270.271	1.249	~~O_3 column 279.9 DU~~
3.066	704.946	270.121	1.239	
3.099	702.000	269.974	1.229	
3.147	697.763	269.671	1.201	
3.262	687.664	268.899	1.127	
3.497	667.380	267.256	0.966	
3.600	658.649	266.536	0.898	
3.700	650.259	265.838	0.833	
3.800	641.950	265.139	0.769	
3.900	633.727	264.440	0.707	
4.000	625.592	263.741	0.645	
4.100	617.538	263.042	0.585	
4.200	609.570	262.342	0.526	
4.300	601.680	261.644	0.468	
4.400	593.871	260.919	0.446	
4.500	586.147	260.186	0.436	
4.600	578.503	259.455	0.425	
4.700	570.935	258.723	0.415	
4.800	563.441	257.991	0.405	
4.900	556.027	257.260	0.395	
5.000	548.693	256.528	0.385	
5.500	513.149	252.871	0.338	
6.000	479.451	249.366	0.261	
6.500	447.548	245.966	0.169	
7.000	417.379	242.567	0.087	
8.000	361.924	235.748	0.027	
9.000	312.542	228.923	$9.69 \cdot 10^{-3}$	
10.00	268.744	222.463	$2.96 \cdot 10^{-3}$	
15.00	123.703	213.438	$4.17 \cdot 10^{-4}$	
20.00	54.6496	209.890	$2.35 \cdot 10^{-4}$	
30.00	10.775	212.607	$6.27 \cdot 10^{-5}$	
40.00	2.488	248.853	$1.41 \cdot 10^{-5}$	
60.00	0.179	239.829	$9.17 \cdot 10^{-7}$	
100.0	$2.77 \cdot 10^{-4}$	213.601	$1.13 \cdot 10^{-10}$	
120.0	$2.38 \cdot 10^{-5}$	378.719	$8.74 \cdot 10^{-13}$	

Tab. A2: Mean atmospheric state of the closure data set: trace gas column amounts. The data set was selected from Zugspitze solar FTIR spectra measured from Dec 2013 – Feb 2014 and contains 52 spectra. Spectra selection criteria are listed in Sect. 3.3.

IWV	2.26 mm
XCO_2	395.3 ppm
XCH_4	1781 ppb
XN_2O	311.8 ppb
O_3 column	279.9 DU

Formatierte Tabelle

Figures

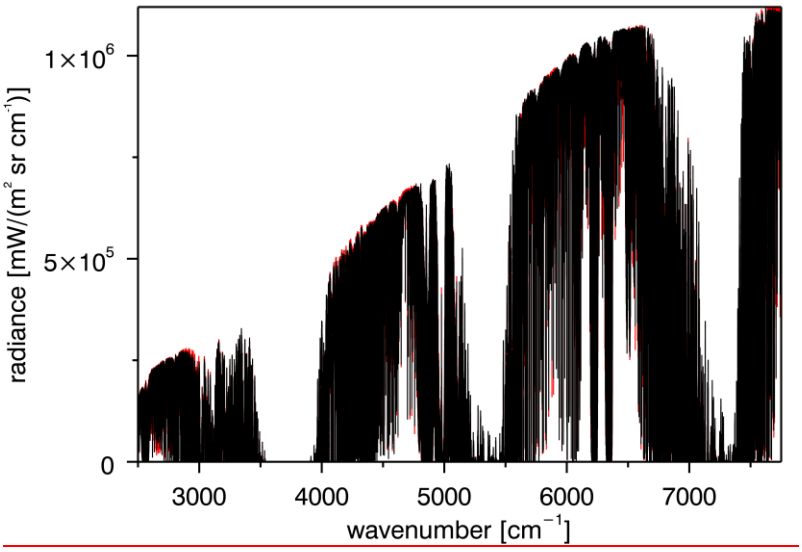

Fig. 1: Mean measured (black) and synthetic (red) radiance spectra for the closure data set selected according to the criteria presented in Sect. 3.3.

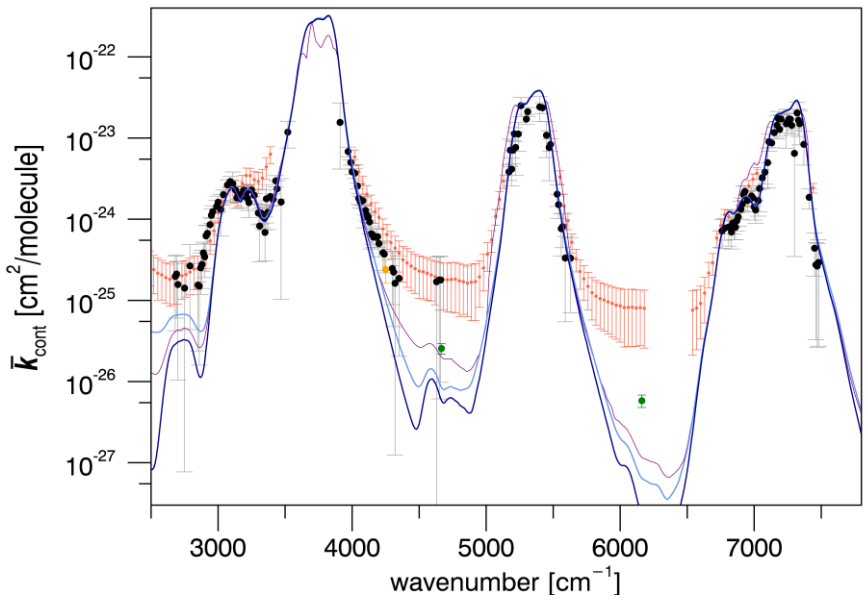

Fig. ~~1~~2: Mean continuum absorption coefficient $\bar{k}_{cont}$ determined in the Zugspitze closure experiment and corresponding 2-σ uncertainties (black). The figure only includes data points for which the measured continuum exceeds the estimated uncertainty. A representation of the full set of measurement results is shown in Fig. B1. Results are compared to the MT_CKD 2.5.2 model (self and foreign continuum: light blue, foreign continuum: dark blue), the BPS-MTCKD 2.0 model (purple line) and the following laboratory studies carried out at room temperature or below: the CRDS measurements of Mondelain et al. 2015 (orange), the calorimetric-interferometric measurements of Bicknell et al. (2006) (green) and the FTIR measurements of Ptashnik et al. (2012; 2013) (red).

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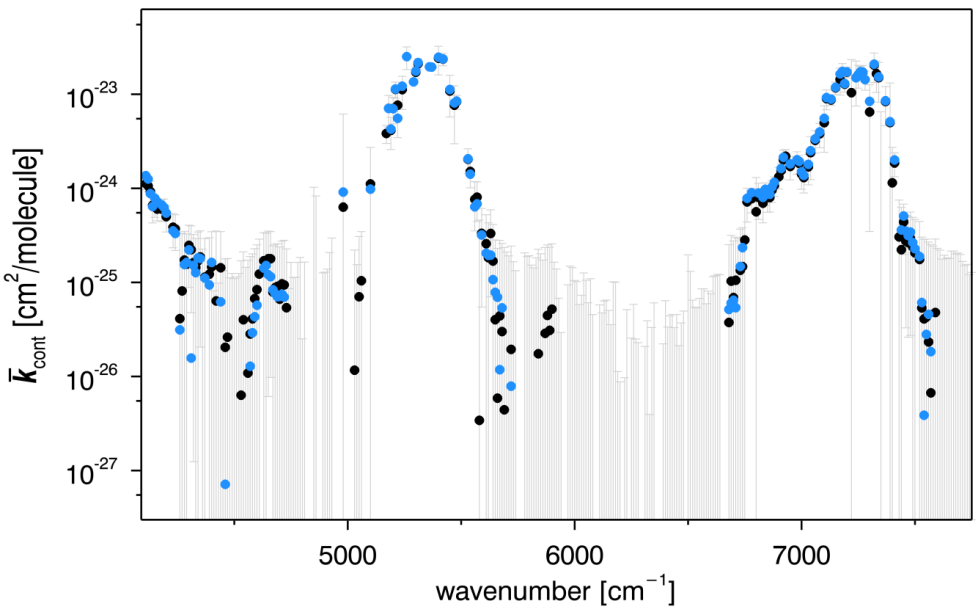

Fig. 3: Mean continuum absorption coefficient derived with the method and data set described in Sect. 3 using the ESS by Menang et al. (2013) (blue data points) and by Kurucz (2005) (black data points). The different ESS sources differ by about 5 % and many solar lines not present in Kurucz (2005) were included in Menang et al. (2013).

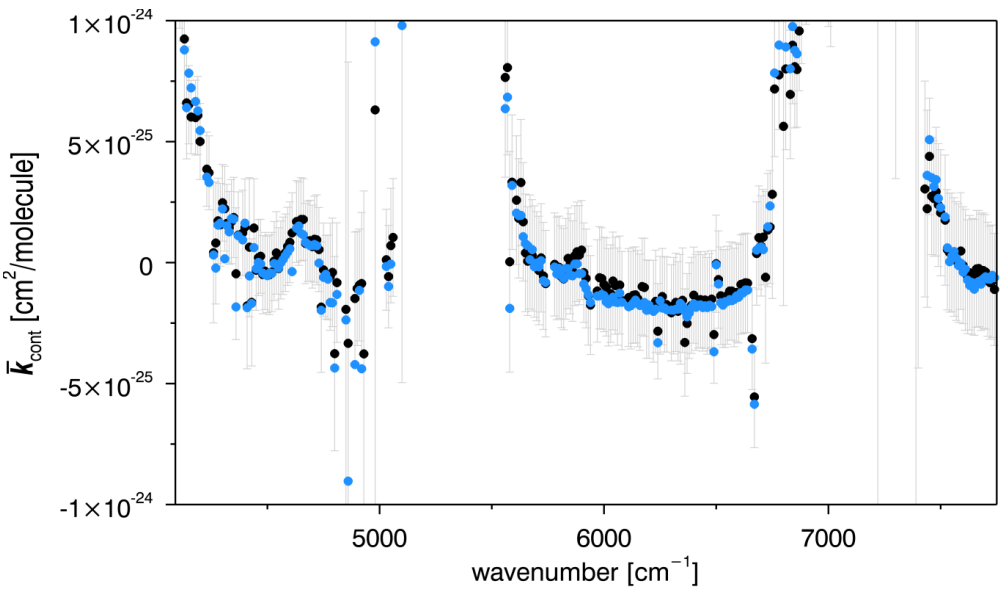

Fig. 4: Same as Fig. 3 but using a linear scale to show the results in the window regions.

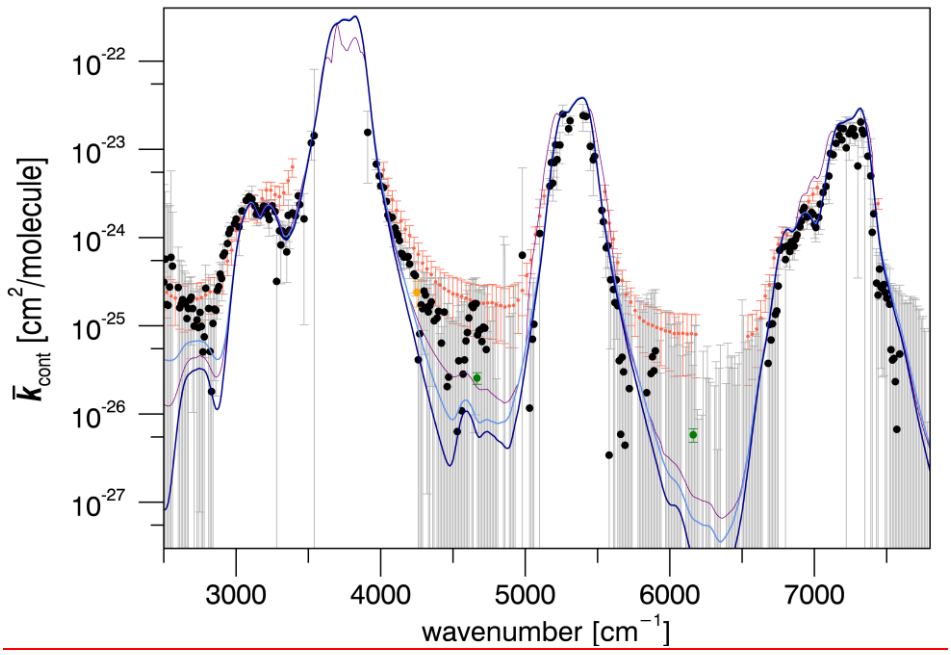

Fig. B1: Mean continuum absorption coefficient $\bar{k}_{cont}$ determined in the Zugspitze closure experiment and corresponding 2-σ uncertainties (black). Results are compared to the MT_CKD 2.5.2 model (self and foreign continuum light blue, foreign continuum dark blue), the BPS-MTCKD 2.0 model (purple line) and the following laboratory studies carried out at room temperature or below: the CRDS measurements of Mondelain et al. 2015 (orange), the calorimetric-interferometric measurements of Bicknell et al. (2006) (green) and the FTIR measurements of Ptashnik et al. (2012, 2013) (red).

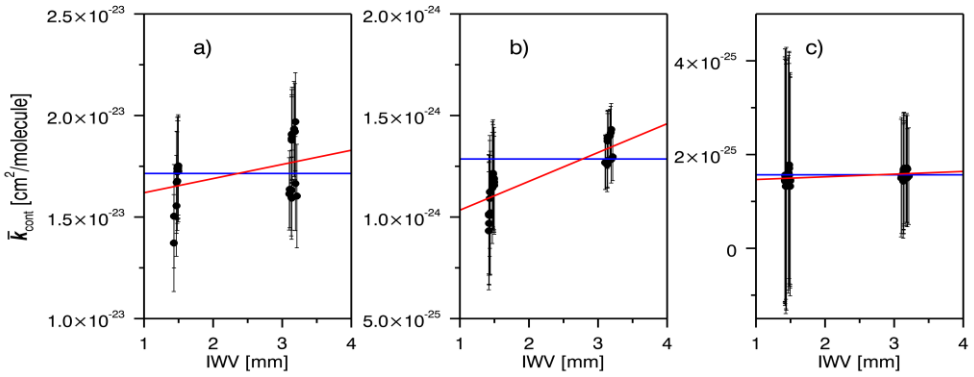

Fig. B2: Examples of the scaling of the mean continuum absorption coefficient $\bar{k}_{cont}$ (black data points) with respect to IWV a) within a water vapor absorption band (4100 cm^{-1}), b) in the wings of an absorption band (4700 cm^{-1}), and c) in a window region (2700 cm^{-1}). The blue line corresponds to the best fit constant scaling, the red line to the best fit linear scaling.

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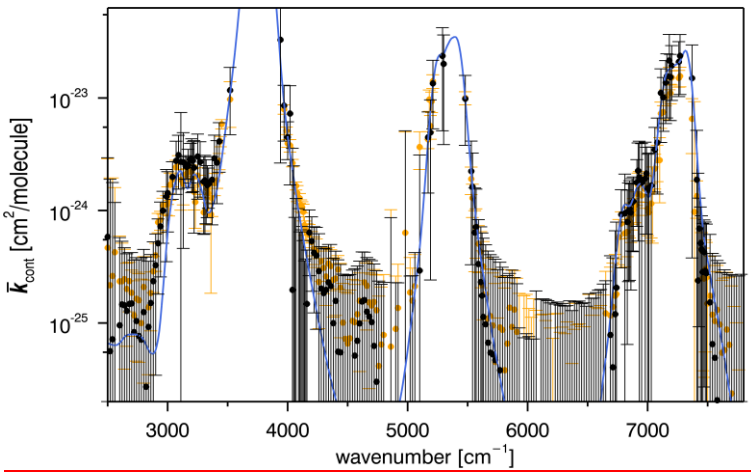

Fig. C1.: Mean continuum absorption coefficient $\bar{k}_{cont}$ determined from 12 December 2013 spectra using the Langley method and corresponding 2 σ uncertainties (black). Results are compared to the calibrated method for the same spectral dataset (orange) and the MT_CKD 2.5.2 model (blue).

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