Final technical report

# Climate-Relevant Gas Absorption Properties from AWARE and Other ARM Spectral Measurements


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## 1. Introduction

The objective of this project has been to use measurements from the Atmospheric Emitted Radiance Interferometer (AERI) deployed at ARM sites to improve our knowledge of uncertain infrared spectroscopic parameters of importance to climate, remote sensing, and data assimilation. A primary focus has been the water vapor continuum in the infrared atmospheric window. Radiation codes used to predict climate and weather base their representation of the water vapor continuum in this window on the MT_CKD model, but in this spectral region the MT_CKD water vapor continuum absorption coefficients were derived almost two decades ago by an analysis of AERI measurements for conditions with a limited range of precipitable water vapor (PWV) and temperature values, leading to uncertainty in the derived coefficients. This project has included a new, comprehensive analysis of all aspects of the water vapor continuum in the atmospheric window -- the self continuum, the foreign continuum, and the self continuum temperature dependence, all resolved spectrally. Having a wide range of PWV amounts and temperatures from deployments of the ARM Mobile Facility for GoAmazon as well as the more than a decade of AERI measurements at SGP has provided an appropriate foundation for such a comprehensive analysis.

This primary objective depends on having an accurate specification of the water vapor field above the AERI instrument during each deployment. Radiosondes provide a solid initial specification of water vapor, but have been shown to be subject to both biases and variability. For two decades, the standard specification of water vapor at ARM sites has involved modification of sonde water vapor profiles such that the profile is consistent with water vapor information obtained from microwave instruments measuring in the vicinity of water vapor absorption lines. Resultingly, a secondary focus of this project has been on the evaluation and improvement of water vapor spectroscopy in the microwave.

We also exploited the low PWV values associated with the RHUBC-II campaign to begin an analysis of the spectroscopic parameters in the key infrared water vapor absorption band ($n_2$) used for remote sensing and data assimilation of water vapor. This will be coupled with a complementary analysis of IASI satellite observations in this band, which is ongoing.

All improved water vapor continuum parameters that have been derived in this project have been implemented in the MT_CKD continuum model, and values that have not yet been finalized will also be included. Improvements will also be utilized by to the Line-By-Line Radiative Transfer Model (LBLRTM), and the fast radiation code RTE+RRTMGP. In addition, the accuracy of a number of standard ARM products will be enhanced by the improvements in spectroscopy resulting from our study, including the MWRRET PWV/LWP retrieval and AERIoe retrievals. RTE+RRTMGP will be implemented in E3SM, CESM, and other atmospheric models, so the result of this project will lead to important improvements in the treatment of radiation in DOE-supported global and regional climate models.

Section 2 of this report details the improvement to the MT_CKD water vapor continuum in the microwave. This improvement led to our undertaking the main focus of our research, the determination of water vapor continuum absorption in the infrared window, which is presented in Section 3. Finally, Section 4 presents information about our work in the water vapor $n_2$ band and other accomplishments of this project.

## 2. Water vapor continuum in the microwave

As mentioned above, all objectives of this project depend on having an accurate specification of the water vapor field for each AERI observation. In previous analyses using ARM observations, measurements from microwave radiometers provided essential information on the water vapor profile. Therefore, a focus of this project was on the evaluation and improvement of water vapor spectroscopy in the microwave (MW).

In the first year of the project, we applied the methodology used in Payne et al. (2011) in an analysis of a 3-year dataset (2005-2007) from SGP to more recent observations by ARM microwave radiometers (MWRs). The sites and time periods of MW observations to be analyzed were decided and the data obtained (along with complementary data, such as radiosondes). For this initial analysis, we applied our methodology to two years of data from SGP (2014, 10/2016-9/2017), one year of data from TWP/Darwin (2015) and one year of data from GoAmazon (2015). We matched up the radiometer observations within a window around sonde launch times, and removed cases not associated with clear skies. Our initial analysis determined that MWR measurements at TWP/Darwin were not consistent with those from SGP and GoAmazon, so the Darwin cases were not included in further analysis.

Using the SGP and GoAmazon MWR dataset, we found that a large revision to the MW water vapor continuum was necessary, with the self continuum requiring a ~50% increase and the foreign continuum a ~25% decrease. This surprising preliminary conclusion was presented at the ASR Science Team meeting and in other presentations in 2018. Even ignoring the GoAmazon observations, the 2-year SGP MWR dataset employed in this analysis was clearly not consistent with the 3-year (2005-2007) SGP MWR dataset used in Payne et al. (2011), despite use of the same instrument and calibration process. It is important to point out the differences between these MWR datasets are within the stated accuracy range of the instrument – our analysis was pushing the instrument's performance to and beyond the typical demands that have been placed on MWR observations, a realization that escaped us on previous similar studies.

Following the determination of the inconsistency between the two SGP datasets and our need for great trust in the dataset we ultimately utilize to determine the MW continuum, we decided to take a careful look at the performance of the MWR across the various sites and years under consideration for our study by comparing the observations as a function of sonde PWV. We found that there were notable differences in SGP observations even for recent years. For example, using only the 2015 SGP MWR paints a far different picture than the tentative conclusion discussed above (i.e. large increase in self and large decrease in foreign). Instead, the existing self coefficient in MT_CKD appeared valid and a small increase in the foreign would have been required. Furthermore, the GoAmazon MWR observations were not sufficiently consistent with the recent SGP observations to allow for them to be jointly utilized in our study.

To determine which, if any, time periods had MWR measurements that we could trust for our analysis required evaluating the consistency of candidate instruments and time periods with other co-located instruments. In particular, we evaluated both channels of the MWR with similar channels of the MWR3C and the 23.8 GHz MWR observations with AERI measurements at 985 $cm^{-1}$, with the more definitive criterion being the agreement with the AERI, an instrument in which we have a great deal of confidence. The comparisons with MWR3C indicated that this instrument at SGP was consistent with the SGP MWR (i.e. BT differences are consistent with those predicted by calculations) for some years, in particular 2014 and 2015, but inconsistent in other years. Also, the MWR3C and MWR at GoAmazon and Darwin were not consistent in any years. We have no reason to trust the MWR3C as a reference instrument for the high level of accuracy we need, so inconsistency of the MWR with this instrument in a year does not rule out the use of the MWR for that year. On the other hand, it is a vote of confidence for the SGP MWR in 2014 and 2015 that it agree with the MWR3C. Given the different levels of agreement between these instruments across this long time series of measurements, we coalesced on a final data set consisting of those clear sky MWR measurements between 2012-2018 that had good agreement with both the contemporaneous AERI and the MWR3C measurements.

Since this MWR dataset did not have sufficient information to determine the temperature dependence of the MW self continuum, before we began our analysis of MWR cases we utilized laboratory studies of the MW self continuum to adjust the temperature dependence of the MT_CKD self continuum. We analyzed the measurements of the self continuum across a range of temperatures presented in Tretyakov et al. (2016), in particular the study of Koshalev et al. (2011), and determined that a ~40% increase in the MW self continuum temperature dependence was needed in the temperature range of greatest applicability in the Earth's atmosphere (275-305K).

Using this adjustment implemented in the AER radiative transfer MonoRTM, we performed calculations of the radiances at the two MWR frequencies, 23.8 and 41.4 GHz, for the cases in our data set. Following the usual approach at ARM sites (as was done in Payne et al. (2011)), each sonde water vapor profile was scaled so that its PWV resulted in agreement between the MWR 23.8 GHz measurement and the corresponding calculation by MonoRTM. In the Payne et al. (2011) study, all properties of the measurement-calculation residuals considered relevant to determination of self and foreign continuum coefficients were combined in a single cost function that was minimized. For the revised analysis performed in this project, separate analyses were performed for each of the key properties – the chi-squared of the (binned by PWV) residuals (Figure 1-top), the coefficient of the quadratic term in the fit of binned residuals vs. precipitable water vapor (PWV) (Figure 1-bottom), and the coefficient of the linear term in this fit (not shown). Figure 1 shows that the lines of minimum value in these two heat diagrams are nearly orthogonal to each other, so their joint minimum occurs in a limited region of the parameter space of foreign and self continuum scale factors. We determined that scale factors of 0.92 (foreign) and 1.25 (self) were the best values to obtain good results for all three of the key properties analyzed.

With these scale factors applied, revised MonoRTM calculations were performed and the resulting residuals calculated. Figure 2 shows that the residuals are small and the quadratic fit to the residuals as a function of PWV is very close to a horizontal line.

The revised MW foreign continuum, self continuum, and temperature dependence of the self continuum were implemented in MT_CKD_3.5. In addition to these modifications, this revised MT_CKD version also had changes to the self continuum and its temperature dependence in the far-infrared spectral region, which had been shown to be necessary in Odintsova et al. (2020). (This aspect of the development of MT_CKD_3.5 is not detailed here.) MT_CKD_3.5 was released in 2020 and is available to the public at https://github.com/AER-RC/MT_CKD. A publication on this study is in preparation (Payne et al., 2021).

**3. Water vapor continuum in the infrared window**

Absorption by the water vapor continuum in the infrared window is a key element in the Earth's radiative balance and for remote sensing of surface temperature. The strength of this absorption has implications for our climate, impacting the magnitude of water vapor feedback to the temperature change due to increases in greenhouse gases as well as the potential for a runaway greenhouse.

The self continuum values in the IR window in the first version of MT_CKD (v1.0) (Mlawer et al., 2012) were fit to the corresponding coefficients derived in an analysis of field measurements (Turner et al., 2004), and no effort was made in the MT_CKD_1.0 foreign continuum development to modify the coefficients in MT_CKD's predecessor model (CKD). Also, the self continuum temperature dependence in this region was also not modified when going from CKD to MT_CKD. After Turner et al (2004), the uncertainty of the window self continuum, which is the dominant source of absorption in this region, was thought to be low. This perspective was subsequently reinforced by the laboratory study of the window self continuum by Baranov et al. (2008), which showed close agreement with MT_CKD_1.0. For these reasons, MT_CKD in the IR window has not changed since the model's initial version.

The apparent consensus, however, was overstated. The window self continuum values in a slightly earlier laboratory study (Cormier et al., 2005) were ~20% lower than in MT_CKD, a result that was not given much credence in Baranov et al. (2008). In addition, Baranov et al. (2008) pointed out their good agreement with pre-1983 laboratory studies by Burch and collaborators, but those results were superseded by a later study by Burch and Alt (1984), which resulted in self continuum coefficients ~20% lower than the earlier lab studies. Even the field study by Turner et al. (2004) was less definitive than had been thought. Although the foreign continuum in this region is much weaker than the self, it is not negligible. A laboratory study (Baranov and Lafferty, 2009) indicated that the window foreign continuum was 2-4 times larger than in MT_CKD. Had Turner et al. (2004) assumed this higher value in their analysis, the derived self continuum values would have been ~10% lower. A higher foreign continuum would also lead to a similar adjustment to the self continuum coefficients derived in another field experiment with results consistent with MT_CKD, that of Han et al. (1997).

For this project's analysis of the window self continuum, we used AERI measurements at SGP from 2016-2018 and from the MAO AMF deployment from 2014-2015. The extensive data set ensures a wide range of PWV values, which is essential for deriving both the self and foreign continuum values. We improved upon the AERI analysis in Turner et al. (2004) by accounting for a persistent warm bias in AERI observations by deriving a fractional "double-pass" obstruction in the AERI aft optics. As in the Turner et al. (2004) study and the MW study described above, we generated scaled versions of radiosonde moisture profiles to be used as input to the radiative transfer calculation, with the scaling based on the MWR observations and the spectroscopy described above. We also generated a second independent set of water vapor profiles from retrievals based on AERI observations on either side of the $CO_2$ $\nu_2$ band (630-700 $cm^{-1}$). These two independent sets of water vapor profiles allow us to determine the uncertainty in the derived continuum coefficients due to the methodology used to specify the water vapor used as input in the model calculations. The temperature profiles used in the calculations were from the sondes, ozone profiles came from MERRA-2, and the profiles of trace gases $CO_2$, $CH_4$, $N_2O$, CFC-11, CFC-12, CCl4, and CFC-22 came from the priors used in retrievals from the Tropospheric Emission Spectrometer (TES). The $NH_3$, abundances used in the SGP calculations came from the US Standard atmosphere and from the tropical standard atmosphere for the MAO calculations.

At each spectral point, a simultaneous linearized least-squares retrieval was performed of scale factors for both the self and foreign continuum, with separate analyses performed for the SGP and MAO observations. This analysis was done independently using the SGP and MAO data sets. The initial guess for the self continuum in the retrieval was the current MT_CKD spectral values, while, for the foreign continuum, both current MT_CKD coefficients and ones that were modified to reflect Baranov and Lafferty (2012) were used as initial conditions. These two independent retrievals allow us to determine the sensitivity of our final result to a change in initial conditions.

The self and foreign scale factors computed by the SGP retrieval were each used to compute continuum absorption coefficients. which were in turn smoothed into a preliminary revised version of MT_CKD. (The retrieval results using the MAO dataset were generally consistent.) Figure 3 shows the revised self continuum, which is significantly lower than MT_CKD_3.5 -- 8-12% at lower wavenumbers than the 10 μm ozone band (750-980 $cm^{-1}$) and 25-30% at higher wavenumbers (1080-1250 $cm^{-1}$). The lower opacity due to the self continuum is counter balanced by the higher level of absorption derived for the foreign continuum compared to MT_CKD_3.5, as shown in Figure 4. The preliminary revised foreign continuum is greater than MT_CKD_3.5 by ~20% below 850 $cm^{-1}$, but is an order a magnitude greater near 1050 $cm^{-1}$ and ~4 times greater near 1200 $cm^{-1}$. For an atmospheric path with high PWV, the total opacity due to the water vapor continuum is less than in MT_CKD_3.5; for low PWV, it is greater.

A change of this magnitude to the absorption in the IR window will have implications for climate prediction and for applications that utilize this spectral region, so we are carefully scrutinizing our results before generating and then publicly releasing a revised version of MT_CKD. We expect to finalize our results in the beginning of 2022.

**4. Water vapor spectroscopy in the $\nu_2$ band, other accomplishments**

*a. Water vapor spectroscopy in the $\nu_2$ band*

We have laid the groundwork for our study of the spectroscopy of the mid-infrared $\nu_2$ band of water vapor, which is often used to retrieve water vapor profile information from satellite observations. For this study, we will use observations from the AERI deployed during the ARM RHUBC-II campaign as well as from the IASI satellite instrument. These complementary data sets may allow us to evaluate both the water vapor continuum and line widths in this region.

As in the studies described above, it is important to accurately specify the water vapor and temperature fields associated with radiometric observations so that corresponding radiative transfer calculations can be confidently compared to the observations, allowing the spectroscopic parameters to be evaluated. For the AERI component of this study, we have repeatedly refined the retrieval of temperature profiles using AERI observations in $CO_2$ absorption bands in an effort to improve or eliminate systematic residuals near the centers of strong lines, features that will inhibit our ability to analyze water vapor line widths in this region. We have had only moderate success in this effort, so, as a result, we will most likely analyze only the water vapor continuum in this region. For the IASI part of this study, which use the same set of cases analyzed in Alvarado et al. (2013), we are performing new temperature and water vapor retrievals to take into account spectroscopic improvements that have been made in the last several years.

Figure 5 presents these complementary observations and their corresponding residuals with respect to LBLRTM in a 50 $cm^{-1}$ spectral region in the P-branch of the water vapor $\nu_2$ band. In the three microwindows highlighted in this figure, which are in spectral regions that are sufficiently transparent so the water vapor foreign continuum can be assessed, the results indicate that this continuum may be overestimated in this region. Similar results are seen throughout the P-branch (1350-1550 $cm^{-1}$).

As above, the spectroscopic improvements resulting from this study will be implemented in MT_CKD and AER's line-by-line models LBLRTM, CLBLM, and MonoRTM.

*b) Other accomplishments*

**Publications -** During this period of performance, we submitted and subsequently revised a manuscript (Mlawer et al., 2019) on our analysis of far-IR and sub-millimeter spectroscopy from the ARM RHUBC-II campaign. We also contributed to a paper (Gordon et al., 2022) describing the new compilation of HITRAN, which includes the improved far-IR and sub-millimeter line widths determined in Mlawer et al., 2019). Our spectroscopic studies in the MW were discussed at a workshop that was summarized in Mattioli et al. (2019). In addition, we contributed to a paper (Pincus et al., 2020) with reference line-by-line calculations that incorporated improvements from studies we performed with ASR/ARM funding.

**Presentations** - We presented posters on this project at the 2019, 2020, and 2021 ASR PI meetings. The PI of this project presented the initial analysis of the MW continuum study at the "Current status of gas absorption knowledge in the sub-millimetre region" workshop in Darmstadt, Germany, in December 2018. A summary of this workshop on which the PI is a co-author was published in BAMS (Mattioli et al., 2019). This work was also presented by the PI at the 2019 International Workshop of Radiative Transfer Workshop for Satellite Data Assimilation in Tianjin, China. The PI presented our far-IR research at the 2020 AGU conference in a talk, "Far-IR Water Vapor Spectroscopy from Analysis of Field Campaign Measurements".

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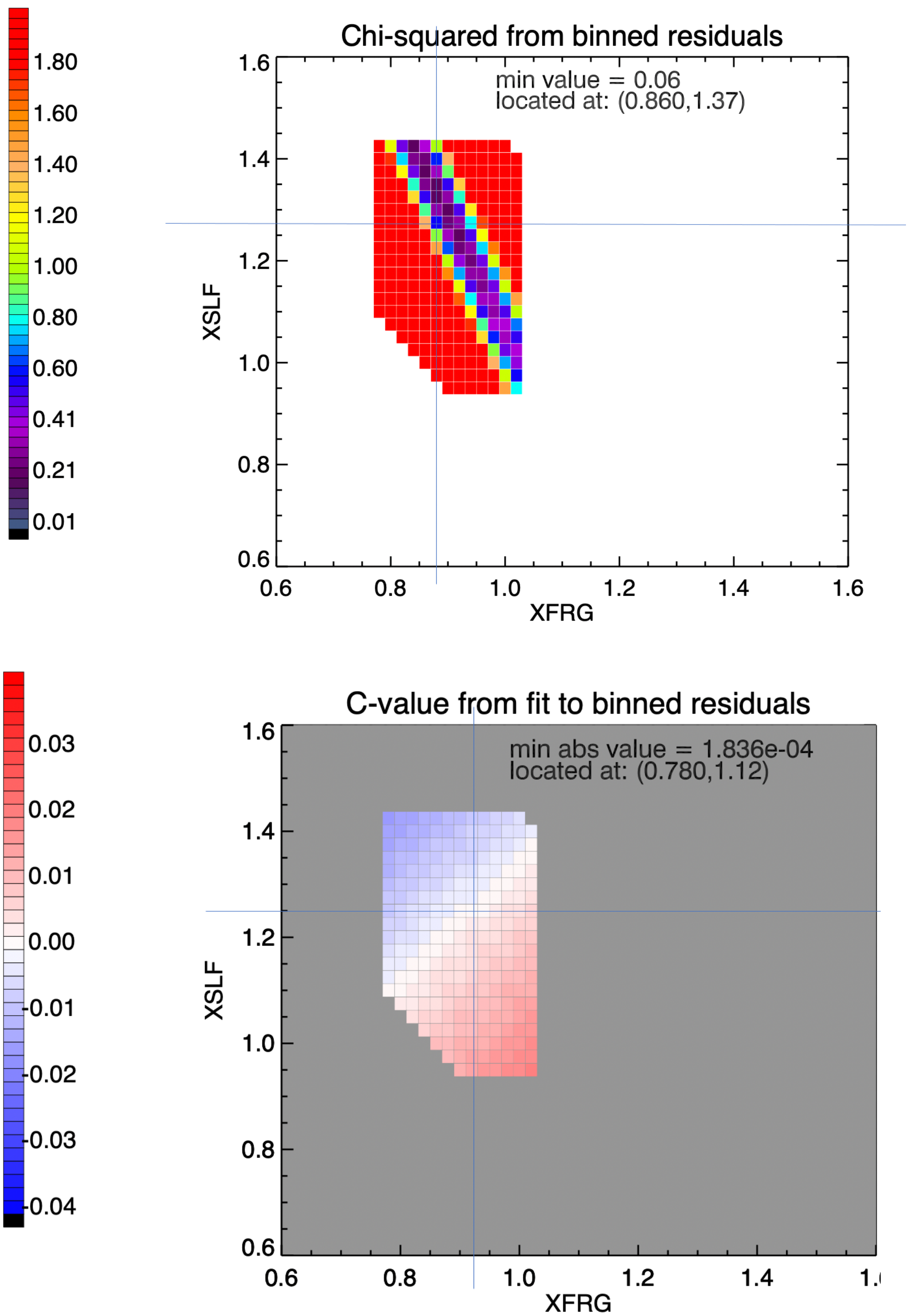


**Figure 1.** (top) Heat diagram of the chi-squared values of measurement-calculation (binned by PWV) residuals at 31.4 GHz for combinations of foreign and self continuum scale factors (based on MonoRTM_5.4, which includes MT_CKD_3.2). (bottom) heat diagram of quadratic term to the fit of residuals vs. PWV. Crosshairs indicate the foreign and self continuum scale factors chosen for MT_CKD_3.5, which is on the axis of minimum values in each heat diagram.

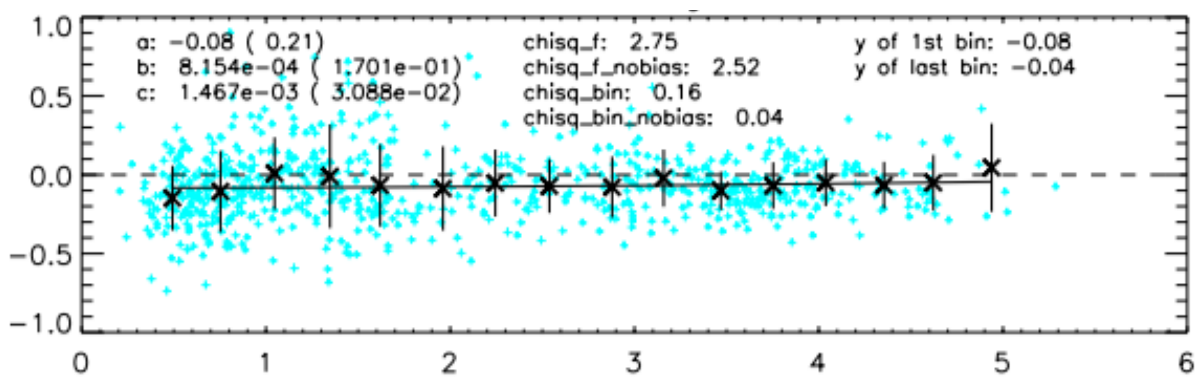


**Figure 2.** For 31.4 GHz, differences between MWR measurement and MonoRTM calculations as a function of PWV bin for the revised water vapor continuum parameters (MT_CKD_3.5).

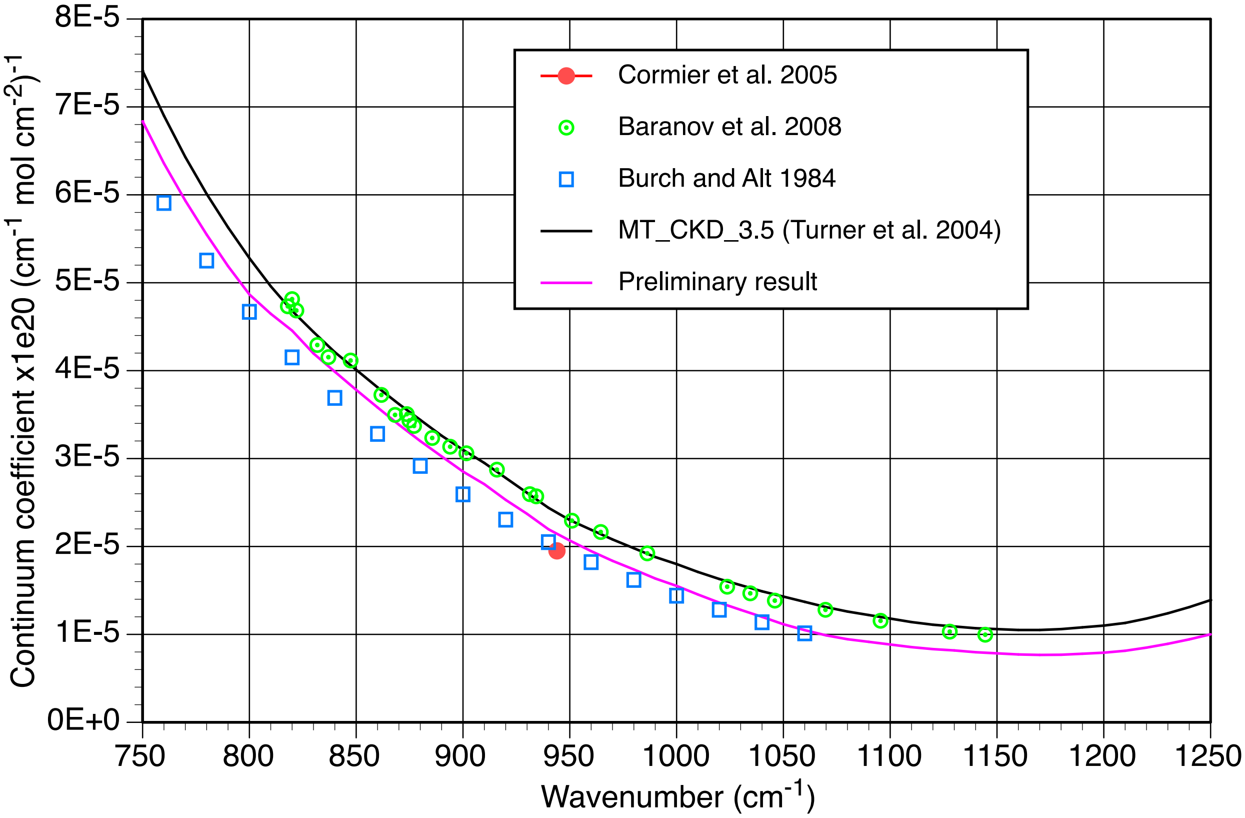


**Figure 3.** Self continuum coefficients (magenta) derived from the retrieval procedure described in the text, along with the current continuum coefficient values (black), and the lab-derived coefficients obtained by Cormier et al. (2005, red), Baranov et al. (2008, green), and Burch and Alt (1984, blue).

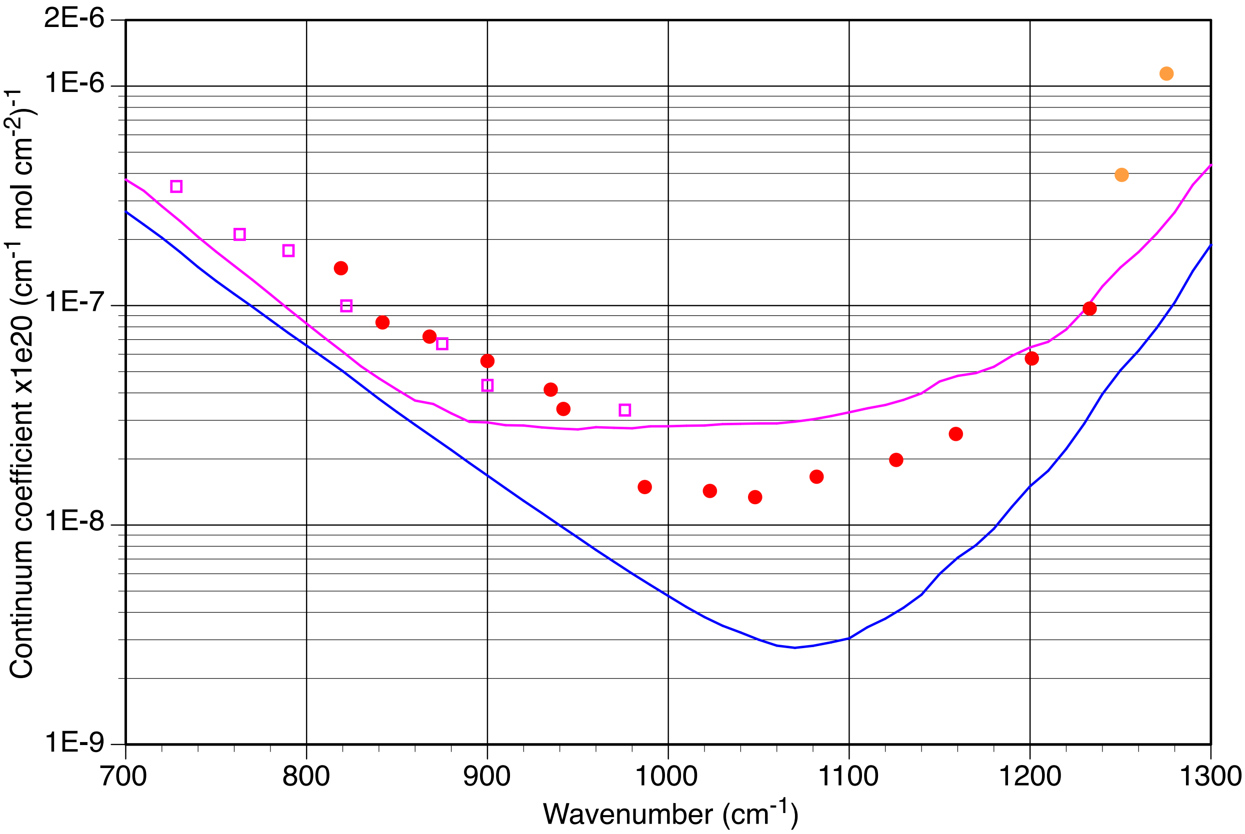


**Figure 4.** Foreign continuum coefficients (magenta) derived from the retrieval procedure described in the text, along with the current (MT_CKD_3.5) continuum coefficient values (blue), and the lab-derived coefficients obtained by Baranov and Lafferty (2012, red), Burch and Alt (1984, pink squares), and Burch (1982, orange).

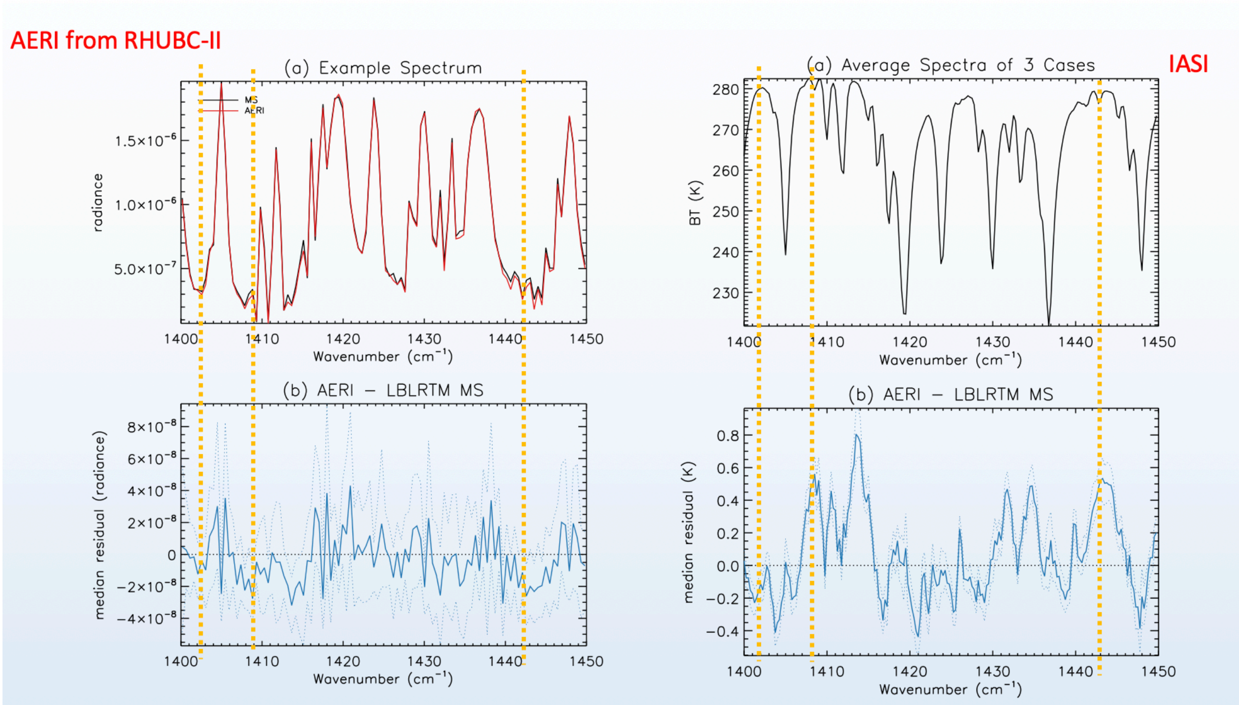


**Figure 5.** (left) (a) Example AERI radiances observed at RHUBC-II from 1400-1450 $cm^{-1}$ and (b) median AERI – LBLRTM residuals in this region from the RHUBC-II data set. (right) (a) Average of three observed IASI radiance spectra from 1400-1450 $cm^{-1}$ and (b) median IASI – LBLRTM residuals for these three cases. The vertical yellow dotted lines are placed in three microwindows of interest (see text).