

General comments:

This manuscript, AMT-2020-94, reports the evaluation of new O₃ xsec data sets (labeled as “BW”) measured in the Hartley and Huggins bands for the use of O₃ profile retrieval from OMI observations. The BW data sets were modeled by using a polynomial in a function of temperature in order to facilitate direct comparison with the current reference data set (“BDM”) and their application to the O₃ profile retrieval. They have found that the new data set, BW, shows a better performance in the retrieval of O₃ profile in terms of less oscillatory features in the retrieved profile and better agreement with the ozonesonde data. We found the manuscript written in a nice and compact manner; the presentation looks consistent. However, we are not convinced that we can agree with the authors’ interpretation of what is described in Sec. 2, which will be detailed below.

This manuscript has shown well that the new dataset, BW, is better than the BDM in the O₃ profile retrievals primarily because of their wider temperature coverage, esp. going down to 194 K critical to the retrievals in the transition layers (UTLS), which was not covered by the BDM dataset in temperature. Therefore, the conclusion of this work has been supported by the results presented in the manuscript. The topics of this paper highly relevant to the scope of AMT, so that we recommend a publication of this manuscript to AMT with a revision or a further clarification Sec. 2. Specific comments and suggestions follow.

Responses to general comments

We would like to thank this reviewer for the constructive comments. We did our best to sincerely reply to 4 comments made by this reviewer.

Specific comments

C1. The authors wrote “Offset corrections were made for each of the 6 temperatures by fitting to the SER dataset since it was measured at higher ozone column density and thus considered more reliable regarding offset”. Does this mean that the BW xsec was normalized to that of SER. Clarify what the corrections factors were and how (and what wavelengths) they were determined. Was this offset considered in the error budgets?

R1. Offset errors in the baseline of the measured spectra cause offset errors in the absorption cross section. Since the column amount of the ozone was limited by the relatively small absorption path of 22.1, the offset error in the ACS was relatively large, up to $2e-22$ cm²/molec. Around 344 nm this amounts to about 20% of the ACS. At 330 nm the offset is about 4%. At 270 nm the offset is about 0.0025%. In order to correct this error fits of the BW ACS to the SER ACS fitting a scalar and an offset were performed in the range 317-350 nm. The offset error in the SER ACS were much smaller due to the significantly longer absorption path (270 cm). The scalar was ignored. The offset was used to correct the entire wavelength range, but it would not have made a difference if we had limited it to the fit range since the offset error influence below 330 nm is negligible. The offset uncertainty was determined from the standard deviation of the fit multiplied with chi since the residuals were not purely noise. The offset uncertainty was $1e-24$ cm²/molec, which is negligible. We think that this discussion is beyond the scope of this paper, which is not intended, for developing/introducing this spectroscopic data, but for applying this dataset on our retrievals. The related discussion will be addressed in a separate paper lead by the author of this dataset, Manfred Birk.

C2. Author wrote, “After offset correction polynomials of 1st order (<270.27 nm) and 2nd order (>270.27 nm) in temperature were fitted for each spectral point to improve the statistical uncertainty” and followed by “Measured cross-sections are typically parameterized quadratically to be applied conveniently at any atmospheric temperatures” using the following equation: $C = C_0 + C_1(T - 273.15) - C_2(T - 273.15)^2$.

C2-1. The agreement between the original data and the fitted data should be inspected or discussed for each of the two data sets, BW and BDM, and discussed. Besides, direct comparison of their original

data sets between BW and BDM (prior to having them fitted to the polynomial), which may be done at $T = 273$ and 295 K provided that their temperature differences, $\Delta = 0.5$ and 0.7 K, respectively, is insignificant, which seems true because the authors argued the dominant coefficient C_0 is almost independent of temperature.

R2-1. As mentioned in Section 2, the temperature correction has already been applied in the BW dataset available to the public. This paper is devoted to atmospheric validation of the BW dataset, rather than presenting the dataset itself. We think that it is out of scope to give a detailed evaluation for the original BW dataset where either offset and temperature correction is turn off because it is not officially published. The detailed views on the original/corrected BW dataset will be provided in another paper written by Birk and Wagner. In the ozone profile algorithm the cross sections parameterized using this quadratic equation are typically used to represent the dependence of cross-section on the atmospheric temperature vertically rather than the interpolated spectrum from original measurements. Therefore, this paper focused on comparing coefficients and the parameterized cross-sections between BDM and BW datasets.

C2-2. We are not sure how well the Eq. (1) could have captured the temperature dependence of the xsec. The xsec can be represented by integrated (line) intensities for the given frequency (wavelength) grid, and the temperature dependence of the line intensities can be modeled by two parameters, i.e., partition function (which we know well for O3) and the lower state energies (which we do not know for the features of this work). Thus, one can simulate the intensity ratio to that at 296 K at various temperature for a few representative cases of the lower state energies, as shown in Fig. X below. As we see, Fig. X is similar to the right panel of Fig. 1, except for one thing that each curve in Fig. X represents different values of the lower state energies, not the wavelength presented in Fig. 1. There is a possibility of having the sampled wavelengths (such as $280, 290, \dots$, in nm) possessing progressively higher value of their (effective) lower state energies more appropriate to assume that each curve in Fig. 1 corresponds to a different of multiple transitions falling into the particular wavelength data point grid (for instance, $280\text{nm} \pm \text{resolution}$ element). This point should be addressed properly to keep naive readers from being misled to think the temperature dependences in Fig. 1 is attributed to the wavelengths.

R2-2. The quadratic equation was first found to represent well the temperature dependence of ozone cross sections in the UV [Paur and Bass, 1985] and has now become the standard approach [Liu et al., 2007;2013; Chehade et al., 2013a,b; Serdyuchenko et al., 2014]. In addition, Fig. X (this reviewer plotted) and Fig. 1 in this paper commonly imply that the dependence of the cross-section on the temperature tends to be linear at shorter wavelengths and slightly non-linear at longer UV wavelengths. Therefore, the quadratic (2nd) polynomials seem to be adequately fit the cross-section measurements. In revised manuscript, this discussion has been better specified by adding “This quadratic equation was first found to represent well the temperature dependence of ozone cross section in the UV (Paur and Bass, 1985) and has now become the standard approach (Liu et al., 2007; 2013; Chehade et al., 2013a;2013b; Serdyuchenko et al., 2014)” after the equation 1. The approach suggested by the reviewers is somewhat similar to pseudolines that is sometimes employed in the parametrizing the IR cross-sections, where temperature and pressure-dependent cross-sections are fit to a HITRAN-like line list where “transitions” do not have quantum mechanical meaning but do reproduce cross-sections. However, this approach is a lot more sophisticated than suggested by the reviewers because there are more than one transitions (with different intensities and lower state energies) that underlie absorption at selected wavelength. This very non-trivial and intense task has never been applied to the electronic spectra yet.

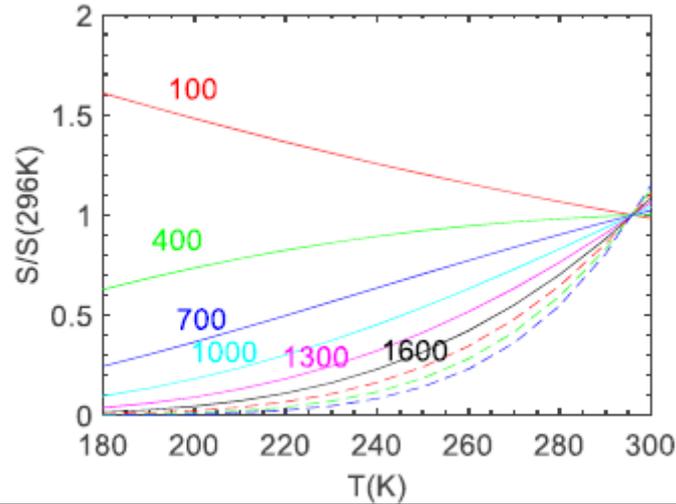
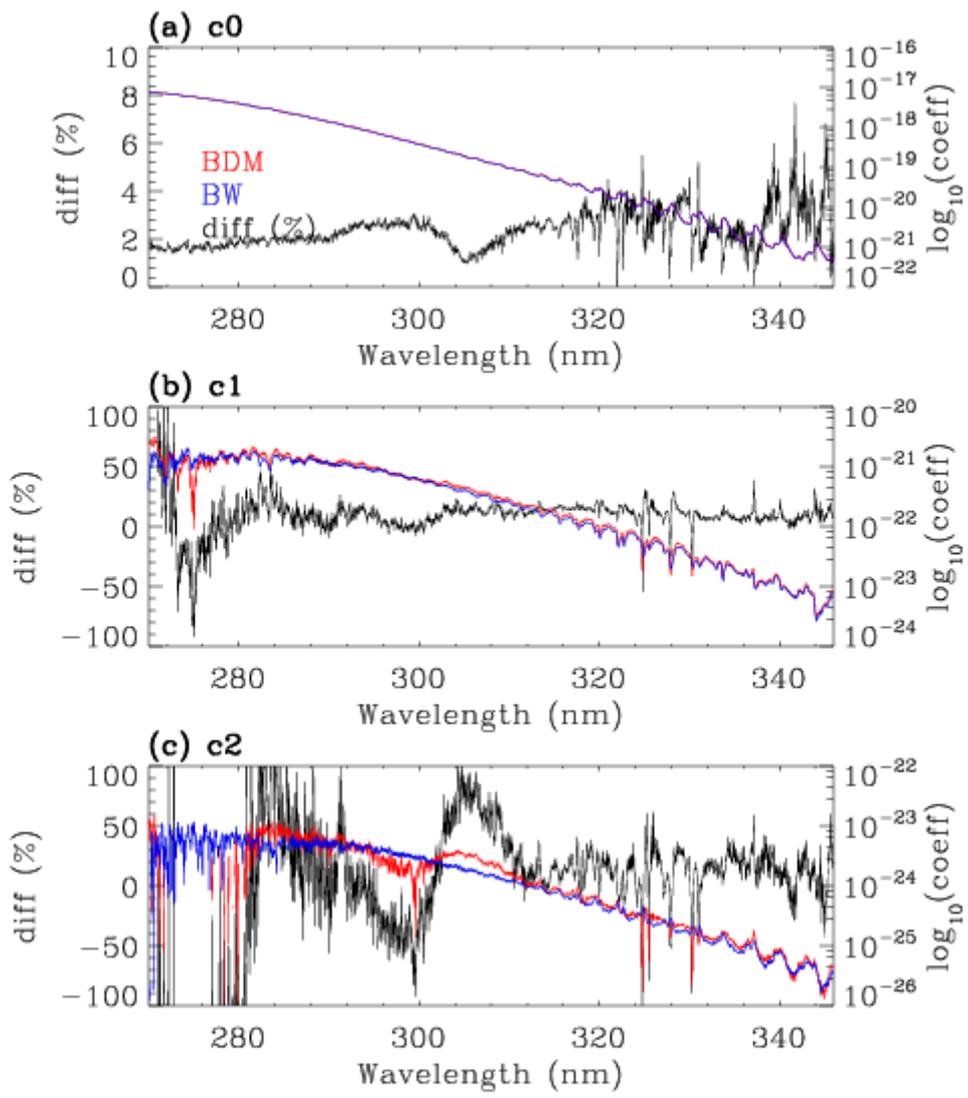


Fig. X $S/S(296K)$ vs. T at a given $E''(\text{cm}^{-1})$
 $= [100, 400, 700, 1000, 1300, \text{etc.}]$

C2-3 For the same reason, Fig. 2 is hard to interpret. The respective comparison of the C1 and C2 for two different data set as a function of nm could be legitimate only when the two data sets are measured at the same resolution because the effective lower state energies mentioned above would be the same. Therefore, the non-wavy feature of C2 for the BW data set would have more to do with the outcome of the resolution choice in the representation by Eq.(1), rather than it is telling the BW data set is superior to the BDM dataset in the temperature consistency. In other words, Fig. 2 shows which data set is better represented by Eq. 1 rather than which data is closer to the truth. This section may stay, but with a specific statement, being provided for the readers on the point made above. The bottom line is that the BW data set is better than the BDM set because of the broader coverage of the measurement temperature, especially covering the temperature critical to UTLS layers, as was properly concluded by the authors in the manuscript.

R2-3. We agree with this comment; it could be not straightforward to compare the coefficients especially C1 and C2 derived from BDM and BW, respectively, due to different spectral resolutions and the strong correlation between C1 and C2 especially when the temperature dependence is weak. However, important insights are obtained from this figure; the comparison of C_0 indicates systematic biases between two datasets, by 2 % on average, with some spikes of up to 8 % at longer UV wavelengths above 315 nm mainly due to the different spectral resolution. The C_1/C_2 characterizes the linear/non-linear dependence of the cross-sections. As shown in Figure 3.c, the quadratic temperature dependence show different behaviors in 290-310 nm, which is significantly correlated with the comparison of cross-section spectrum shown in Figure 4.



Revised Figure 4.