Journal of Quantitative Spectroscopy & Radiative Transfer ∎ (∎∎∎∎) ∎∎∎–∎∎∎

Contents lists available at SciVerse ScienceDirect

Journal of Quantitative Spectroscopy & Radiative Transfer

journal homepage: www.elsevier.com/locate/jqsrt

The impact of using different ozone cross sections on ozone profile retrievals from OMI UV measurements

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ARTICLE INFO

Article history:
Received 2 March 2013
Received in revised form
5 June 2013
Accepted 10 June 2013

Keywords:
Ozone cross section
Ozone profile retrieval
OMI
UV
Ozonesonde
Validation

ABSTRACT

We compare three datasets of high-resolution O_3 cross sections and evaluate the effects of using these cross sections on O_3 profile retrievals from OMI UV (270–330 nm) measurements. These O_3 cross sections include Brion–Daumont–Malicet (BDM), Bass–Paur (BP) and a new dataset measured by Serdyuchenko et al. (SGWCB), which is made from measurements at more temperatures and in a wider temperature range than BDM and BP, 193–293 K. Relative to the BDM dataset, the SGWCB data have systematic biases of −2 to +4% for 260–340 nm, and the BP data have smaller biases of 1–2% below 315 nm but larger spiky biases of up to ±6% at longer wavelengths. These datasets show distinctly different temperature dependences. Using different cross sections can significantly affect atmospheric retrievals. Using SGWCB data leads to retrieval failure for almost half of the OMI spatial pixels, producing large negative ozone values that cannot be handled by radiative transfer models and using BP data leads to large fitting residuals over 310–330 nm. Relative to the BDM retrievals, total ozone retrieved using original SGWCB data (with linear temperature interpolation/extrapolation) typically shows negative biases of 5–10 DU; retrieved tropospheric ozone column generally shows negative biases of 5–10 DU and 5–20 DU for parameterized and original SGWCB data, respectively. Compared to BDM retrievals, ozone profiles retrieved with BP/SGWCB data on average show large altitude-dependent oscillating differences of up to ±20–40% biases below ~20 km with almost opposite bias patterns. Validation with ozonesonde observations demonstrates that the BDM retrievals agree well with ozonesondes, to typically within 10%, while both BP and SGWCB retrievals consistently show large altitude-dependent biases of up to ±20–70% below 20 km. Therefore, we recommend using the BDM dataset for ozone profile retrievals from UV measurements. Its improved performance is likely due to its better characterization of temperature dependence in the Hartley and Huggins bands.
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1. Introduction

The quality of spectroscopic ozone cross sections, including wavelength and radiometric accuracy, is critical to ultraviolet (UV) ozone retrievals using the Hartley and Huggins bands, especially to ozone profile retrievals. The ad-hoc commission ACSO ("Absorption Cross Sections of Ozone") mandated by the WMO and the International Ozone Commission (IO3C) reviews the most appropriate absorption cross sections to be recommended for use in atmospheric ozone measurements, with a current focus on the Huggins bands. The current recommended ozone cross sections are those published by Bass and Paur (BP, [1,2]). ACSO discussed the implications of a possible switch from BP to the laboratory measurements of the French group from GSMA Reims: Daumont, Brion, Malicet (BDM) [3–6] during its first phase activities (2009–2011). Recently a new and more comprehensive laboratory data set (including a larger temperature range) was measured by

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0022-4073/$ - see front matter Published by Elsevier Ltd.
http://dx.doi.org/10.1016/j.jqsrt.2013.06.006

Serdyuchenko et al. (submitted for publication) at the Institut für Umweltphysik, Universität Bremen [7,8] (http://igaco-o3.fmi.fi/ACSO/cross_sections.html, it is abbreviated as the SGWCB dataset) and ACSO has requested us to investigate whether it could improve atmospheric ozone remote sensing.

Liu et al. [9] investigated the effects of different ozone cross sections on Global Ozone Monitoring Experiment (GOME) ozone profile retrievals using fitting windows of 290–307 nm and 325–340 nm. BP, BDM, and GOME flight model (GFM) cross sections were tested and BDM cross sections were recommended to be used for ozone profile retrievals from UV measurements due to much smaller fitting residuals and better agreement with ozonesonde observations. Liu et al. [10] adapted the GOME algorithm to OMI using the fitting window 270–330 nm. Since OMI retrievals use the whole spectrum from 270 to 330 nm without a significant spectral gap, we evaluate the use of SGWCB data on OMI retrievals together with the BP and BDM data in this study.

This study is organized as follows: Section 2 compares the quadratic coefficients in the parameterization of temperature dependence. Section 3 compares one orbit of OMI retrievals using these cross sections, and Section 4 validates various retrievals against ozonesonde observations. Section 5 presents a summary and discussion of the study.

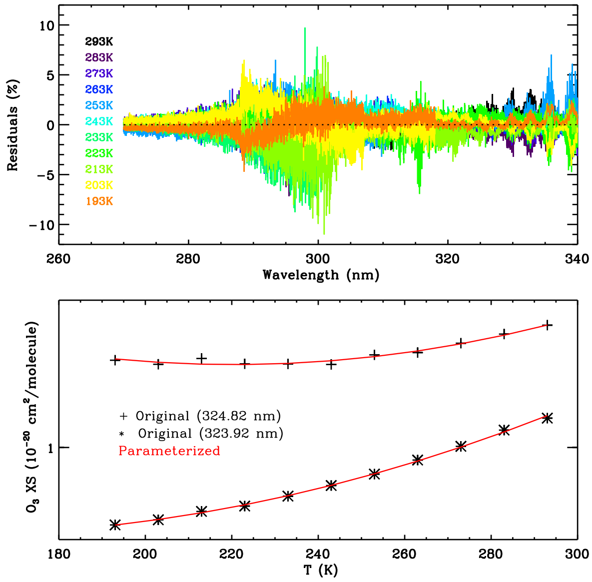

Fig. 1. Upper panel: Residuals in ozone cross sections at different temperatures after using nonlinear least squares fitting to derive quadratic temperature-dependent coefficients from SGWCB O_3 cross sections. Lower panel: Comparison of original and parameterized SGWCB O_3 cross sections at two wavelengths in the Huggins band.

2. Comparison of quadratic parameterization

For convenience of use in atmospheric ozone retrievals that cover a broad temperature range, measured ozone cross sections C are often parameterized quadratically using the following equation:

$$C = C_0 + C_1(T - 273.15) + C_2(T - 273.15)^2 \quad (1)$$

The BDM cross sections were parameterized by Liu et al. [9] using nonlinear squares fitting. The BP parameterization is taken from the original data except that the wavelengths were converted to vacuum wavelengths and shifted by +0.015 nm [11]. Serdyuchenko et al. [8] also provided parameterized O_3 cross sections but only for 300–380 nm. In order to use SGWCB data for our OMI ozone profile retrieval (270–330 nm) [10], we parameterized the data as in Liu et al. [9]. Fig. 1 shows the residuals between the original and fitted O_3 cross sections at different temperatures in the spectral region 260–340 nm and two examples of the fittings (local minimum and maximum) in the Huggins bands. As seen from the differences of $>3\%$ at some temperatures and wavelengths, the parameterization substantially differs from the original data.

Fig. 2 compares the parameterized coefficients for O_3 cross sections in the 260–340 nm range and Fig. 3 shows their relative differences for C_0. The coefficients for SGWCB O_3 cross sections parameterized by SAO are almost the same as those by Bremen, suggesting that the parameterization approaches are very similar. C_0 is similar for these cross sections, with typical biases within 4%. The SGWCB cross sections show biases of –2 to +4% relative to BDM data, and the BP data show smaller 1–2% biases below ~315 nm but larger spiky biases of up to ±6% above 315 nm. For C_1 and C_2, there are distinct differences among these cross sections, indicating different temperature dependences. The BDM cross sections are generally smoother while the SGWCB data show more fine structure and BP data show large oscillating structures, especially in C_2.

3. Effects on OMI O_3 profile retrievals

To evaluate the effects of using different cross sections on ozone profile retrievals, we compare one orbit of OMI retrievals using our ozone profile retrieval algorithm, which was described in detail in Liu et al. [10]. Ozone profiles are retrieved at 24 layers (~2.5 km thick for each layer) from the surface to ~60 km from OMI reflectance spectra in the spectral regions 269–309 nm in OMI UV 1 channel and 312–330 nm in OMI UV2 channel using the optimal estimation technique. Gaussian slit widths are pre-determined for each spectral region by cross-correlating OMI solar irradiance spectra with a high-resolution solar reference spectrum and are used to convolve the high-resolution ozone cross sections. The algorithm fits constant wavelength shifts between radiance and irradiance and between radiance and ozone cross sections in each spectral window. Several changes were made to the algorithm in Liu et al. [10]. Instead of using the altitude-based ozone profile climatology of McPeters et al. [12], we switched to using the recently developed tropopause-based ozone profile climatology [13] as a priori to constrain the retrievals. To account for temperature dependence of ozone absorption, daily NCEP (National Centers for Environmental Prediction) temperature fields are interpolated to the OMI overpass at ~13:45 local solar time at the equator, but switched from the NCEP reanalysis data at 2.5°-longitude × 2.5°-latitude resolution to NCEP Global Forecast System (GFS) final operational analysis

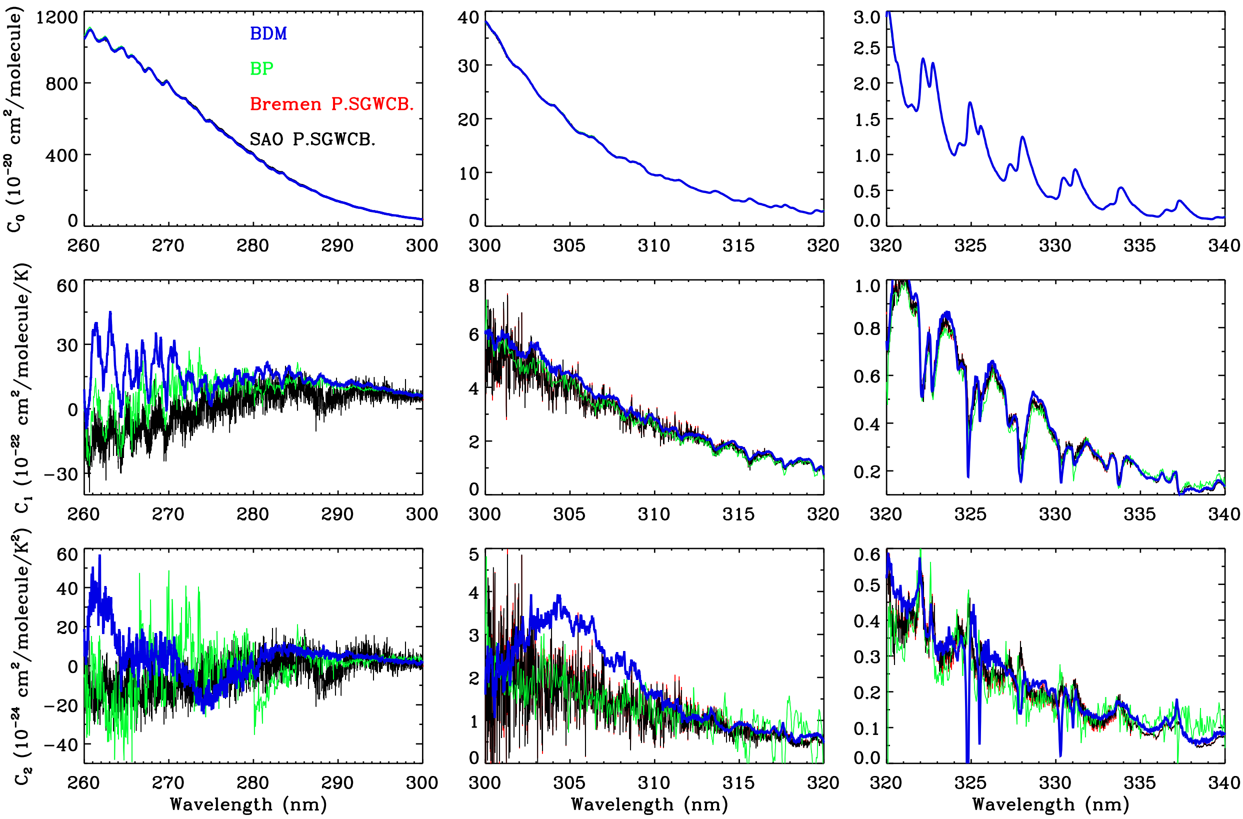

Fig. 2. Comparison of the quadratic coefficients of the parameterization of temperature dependence in ozone cross sections, including SGWCB data parameterized by SAO and Bremen, named SAO P. SGWCB and Bremen P. SGWCB respectively, BDM, and BP data.

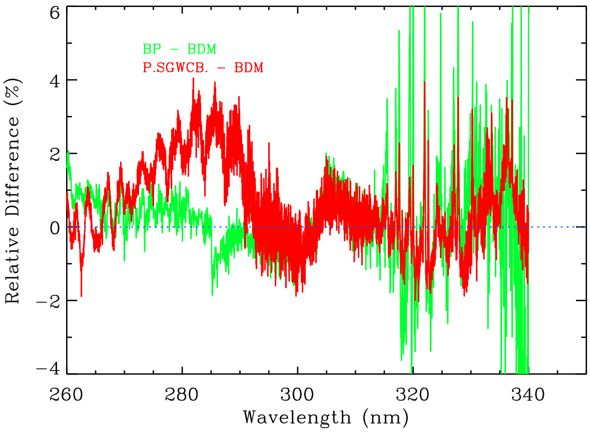

Fig. 3. Relative differences in C_0 between BP/SGWCB and BDM cross sections.

data at $1^{\circ} \times 1^{\circ}$ resolution (http://rda.ucar.edu/datasets/ds083.2/). To conduct a fair comparison, we have turned off soft calibrations in Liu et al., which are derived using BDM cross sections. For SGWCB O_3 cross sections, we tested both the original data (linearly interpolated to atmospheric layer temperature if it is between 193 and 293 K and extrapolated to temperatures outside the range) and parameterized data, since the parameterization

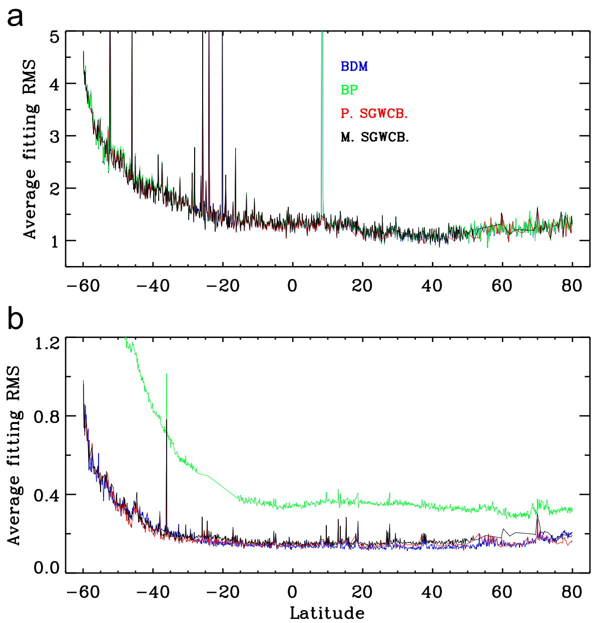

Fig. 4. Average fitting RMS (ratio of fitting residuals to OMI measurement errors) in two fitting channels (269–309 nm, 312–330 nm) using different ozone cross sections, as a function of latitude for the nadir position of orbit 15939 on 14 July 2007. P. SGWCB/M. SGWCB indicate parameterized/original SGWCB O_3 cross sections.

Please cite this article as: Liu C, et al. The impact of using different ozone cross sections on ozone profile retrievals from OMI UV measurements. J Quant Spectrosc Radiat Transfer (2013), http://dx.doi.org/10.1016/j.jqsrt.2013.06.006

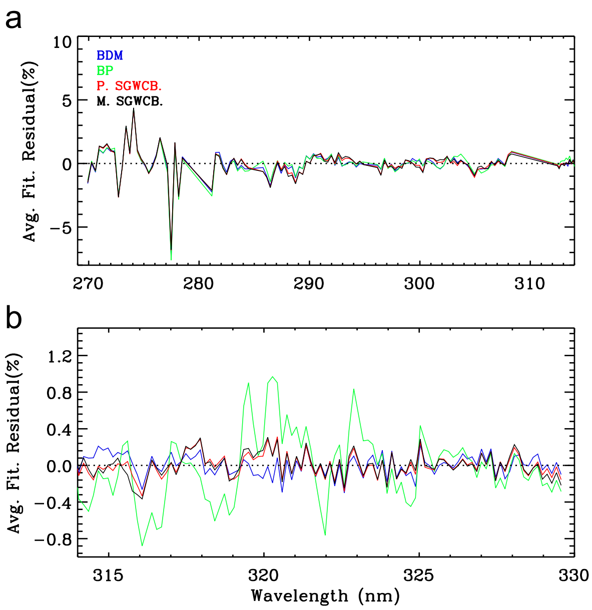

Fig. 5. Average fitting residuals (%) in two fitting windows using different ozone cross sections, as a function of wavelength for the same orbit as in Fig. 4.

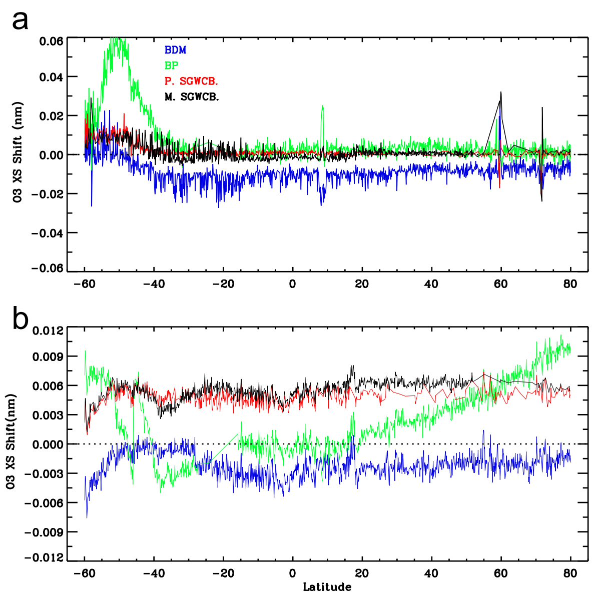

Fig. 6. Same as Fig. 4 except for fitted wavelength shifts between radiances and ozone cross sections.

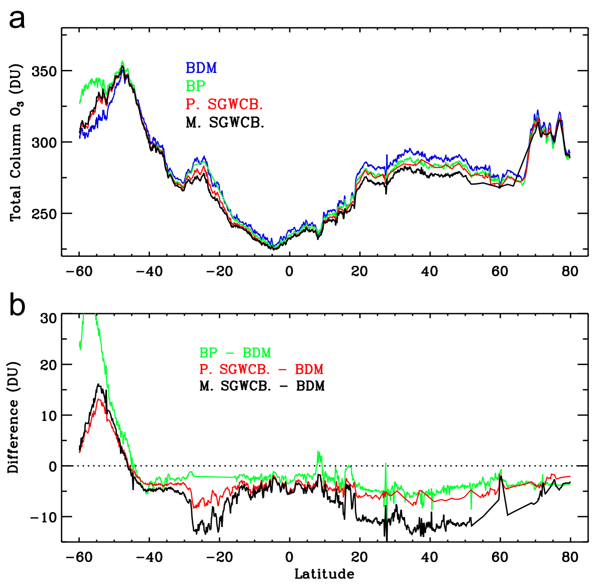

Fig. 7. Comparison of (a) retrieved total column ozone using different cross sections for the same orbit and (b) their differences relative to BDM-derived values.

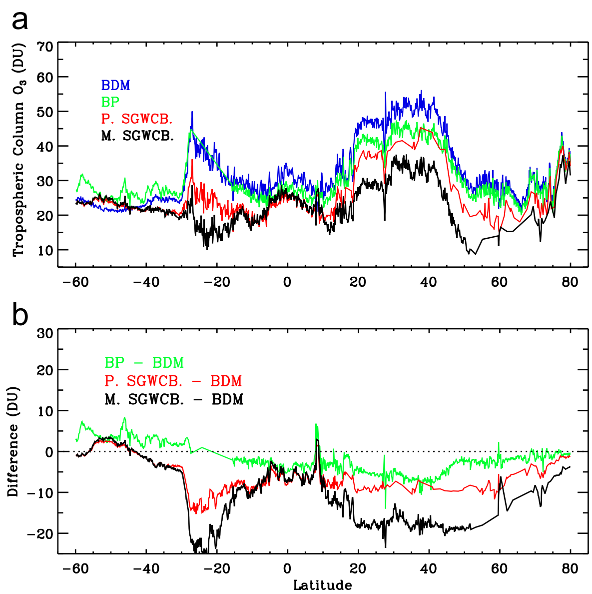

Fig. 8. Comparisons as in Fig. 7 for tropospheric column ozone.

of temperature dependence shows some large residuals (Fig. 1).

Figs. 4–9 compare the average fitting RMS, (Root Mean Square of the ratio of fitting residuals to OMI measurement errors) fitting residuals, wavelength shifts between O_3 cross sections and radiances, total ozone column, tropospheric ozone column, and ozone profile. There are significant differences in the number of converged pixels: 1451, 1265, 816 and 633 pixels for BDM, BP, original and parameterized SGWCB cross sections, respectively. Most of the failed retrievals are due to retrieving large negative values at one or more layers, a situation which cannot be handled by the radiative transfer model. Of all the converged pixels, the average fitting RMS and average fitting residuals show similar behaviors in OMI channel 1, but these are significantly larger for BP O_3 cross sections in channel 2, supporting previous conclusions [10] that the BP data are noisier (in value and/or wavelength precision) than the other two datasets. Shifts in BDM and SGWCB cross sections do not show significant latitudinal dependence except from 40°S to 60°S. For BDM, the mean shift is -0.009 ± 0.006 nm in UV1 and -0.002 ± 0.001 nm in UV2. For SGWCB, the mean shift is 0.0015 ± 0.004 nm in UV1 and 0.005 ± 0.001 nm in UV2. The SGWCB dataset thus has

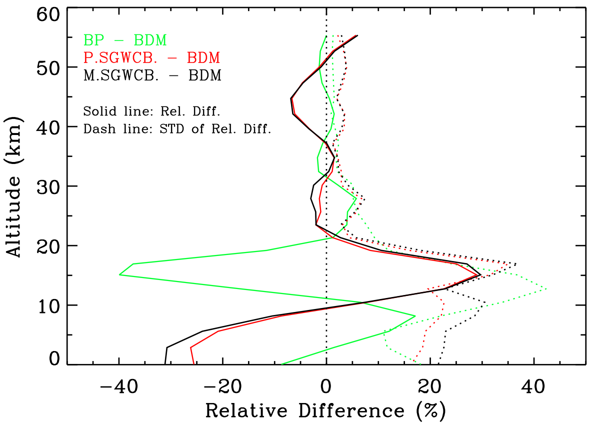

Fig. 9. Mean biases (solid lines) in ozone profiles retrieved using different ozone cross sections and standard deviations (dotted lines) of their differences.

relative wavelength shifts of ~0.01 nm in 269–309 nm and ~0.007 nm in 312–330 nm compared to the BDM dataset. The fitted wavelength shifts for the BP dataset show significant latitudinal dependence in both channels, which suggest that the BP wavelength position might have wavelength-dependent errors.

The selected OMI orbit overpasses the Antarctic, South America, the Eastern Pacific Ocean, North America, and the Arctic. Both total and tropospheric ozone show some strong latitudinal dependence. The total ozone peaks outside the southern polar vortex region at ~350 DU, generally decreases with latitude in the southern hemisphere, minimizes at ~225 DU in the tropics, then generally increases with latitude in the northern hemisphere. There are local maxima at southern and northern subtropics and middle latitudes likely due to enhanced tropospheric ozone. The tropospheric ozone column is generally small except for enhancement in the subtropics and middle latitudes. The retrieved ozone differences between different cross sections also show significant latitudinal dependence due to the interaction between differences in cross sections, including their temperature and wavelength dependences, and variation of temperature, measurement and a priori constraints, and retrieval sensitivity along the orbit.

In the total ozone comparison, retrievals using parameterized SGWCB/BP cross sections typically agree well with BDM retrievals, mostly with negative biases of less than 5 DU, except at south of 50°S where BP retrievals show large positive biases of up to 30 DU and SGWCB retrievals show positive biases of up to 10 DU. Retrievals using the original SGWCB cross sections show large negative biases of 5–10 DU at most latitudes and positive biases of up to 15 DU south of 50°S relative to the BDM retrievals. In general, the total ozone comparison between BDM and BP shows the best agreement, except south of 50°S. The atmospheric effective temperature (temperature weighted by ozone profile) increases sharply from ~205 K to ~225 K from 60°S to 20°S, does not vary much from 20°S to 30°N, and then gradually increases to 235 K from 30°N to 80°N. The variation of effective temperature is anti-correlated with the ozone differences, with a correlation of ~−0.70 for all cross sections. The total ozone differences can thus be well explained by the temperature variation and difference in temperature dependence of the cross sections.

In the tropospheric ozone comparison, there are generally larger absolute differences than for the total ozone comparison except at southern middle and high latitudes, and thus there are much more significant relative differences for the typical tropospheric ozone columns of 10–60 DU. Like the total ozone comparison, the BP retrievals generally agree better with the BDM retrievals than with the parameterized and original SGWCB retrievals. The BP/BDM biases are typically within ~5 DU. The parameterized and original SGWCB retrievals typically show larger negative biases of 5–10 DU and 5–20 DU, respectively, except for southern middle and high latitudes.

Fig. 9 shows the mean relative differences and standard deviations of retrieved ozone profiles using different cross sections relative to BDM retrievals. The relative differences between BDM and other O_3 cross sections are generally within 10% above 20 km. However, there are large altitude-dependent oscillating differences of up to ±20–40% at lower altitudes for both BP and SGWCB retrievals relative to BDM retrievals. Interestingly, the BP biases show opposite behavior of the SGWCB biases: BP retrievals show negative biases of up to 40% at ~15 km and positive biases of up to 20% below 10 km, while SGWCB retrievals show positive biases of up to 30% at ~15 km and negative biases of up to ~30% below 10 km. Differences in the lowermost troposphere (the lowest 5 km, including the planetary boundary layer) are within 10% for BDM but 20–30% for the two SGWCB cases.

To better understand the ozone retrieval differences and their linkage to temperature dependence, we perform retrievals for more orbits (nadir position only, 1 orbit for each month in 2007) that are close in ground track. Fig. 10 shows the mean O_3 differences using different cross sections relative to using BDM cross sections at layers around 5, 15, and 42 km, where there are large O_3 differences seen from Fig. 9, as a function of month for the latitude band of 60°N–40°N along with the time series of mean temperature at each altitude. Table 1 shows the correlations between mean ozone retrieval differences and average temperature at a given altitude range for various latitude bands. We clearly see strong negative correlation of up to 0.7–0.95 between ozone differences and temperatures for both BP and SGWCB cross sections at ~5 km and for SGWCB cross sections at ~42 km when there are large temperature ranges or seasonal variations of temperature (e.g., middle latitudes). Since ozone information at ~5 km mainly comes from the spectral range 300–330 nm, the strong negative correlation suggests that the ozone differences can be well explained by the differences in temperature dependence between different cross sections in this spectral region. Since ozone information at ~42 km mainly originates from shorter wavelengths below 290 nm, it suggests that there are significant differences in temperature dependences below 290 nm between SGWCB and BDM cross sections. At ~15 km, the temperature ranges are relatively smaller and the correlations are generally less significant except for correlation of 0.6–0.78

for the BP cross sections at some latitude bands. Other factors including the vertical smoothing due to the interaction between cross section differences and a priori constraints around the upper troposphere and lower stratosphere may significantly contribute to the large mean differences around 15 km.

4. Validation with ozonesonde observations

To validate retrievals using different cross sections, we compare them against collocated ozonesonde observations (within ± 6 h, where the OMI footprint includes the ozonesonde station) for 2004–2008 at five stations. Ozonesonde profiles are degraded with retrieval averaging kernels to the OMI retrieval resolution. Fig. 11 shows the mean biases and standard deviations at each station in decreasing latitude. As for mean biases, the BDM retrievals generally show good agreement with ozonesonde observations, to within 10%, without significant altitude dependence. The BP and SGWCB retrievals consistently show large altitude-dependent oscillating biases of up to $\pm$20–70% below 20 km, with altitude dependent patterns similar to those in Fig. 9. Generally, the BP/SGWCB relative differences are larger at lower latitudes due to the smaller amount of partial ozone column at each layer. The standard deviations for different cross sections are similar at the two tropical stations, but the BP/SGWCB standard deviations are generally much larger at the other three stations. One exception is at Wallops Island, where the parameterized SGWCB retrievals show similar values as the BDM retrievals, but note that the number of collocations are about half of those using other cross sections since many more of the parameterized SGWCB retrievals did not converge.

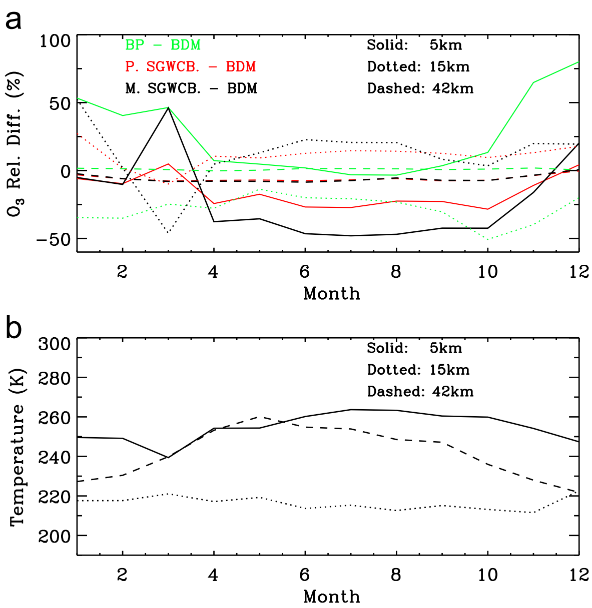

Fig. 10. (a) Average relative differences in retrieved ozone at layers around 5 km, 15 km, and 42 km using BP, P. SGWCB and M. SGWCB cross sections relative to using BDM cross sections vs. month for the latitude band of 60°N–40°N. (b) Corresponding average temperatures at different altitudes vs. month.

5. Summary and discussion

To evaluate the quality in the UV of the new laboratory high-resolution ozone cross section dataset measured by Serdyuchenko et al. [8] for ozone profile retrievals we first quadratically parameterized its temperature dependence and compared it to the other parameterized O_3 cross section datasets, BDM and BP. The parameterization diverges from the original data with large differences, $>3\%$ at some temperatures and wavelengths. Relative to the BDM dataset, the SGWCB data show systematic biases of –2 to +4% in C_0 in the 260–340 nm range and the BP data show smaller biases of 1–2% below 315 nm but larger spiky biases of up to $\pm 6\%$ at longer wavelengths. The differences in C_1 and C_2 indicate distinctly different temperature dependence among these 3 datasets; the SGWCB data show more small high-frequency structures in C_1 and C_2 than do the BDM and BP data.

We then compared ozone profile retrievals from one orbit of OMI UV (270–330 nm) measurements with four cross section datasets: parameterized BDM, BP, SGWCB and original SGWCB (with linear interpolation/extrapolation) datasets and found huge impacts on the retrievals. The use of SGWCB data leads to retrieval failure for about half of the pixels, due to retrieving large negative ozone values at some layers, a situation than cannot be handled by radiative transfer calculations. The three datasets give similar fitting residuals except that the BP residuals are much larger from 312 to 330 nm as a result of the BP

Table 1
Average temperature range in selected days (1 day per month) and the correlation between average temperature and mean relative O_3 differences using different cross sections relative to using BDM cross sections for layers around 5, 15, and 42 km and for various latitude bands.

Latitude bands	Temp. range (K)			Correlation between O_3 rel. diff. and temperature								
				BP–BDM			P.SGWCB–BDM			M.SGWCB–BDM		
	5 km	15 km	42 km	5 km	15 km	42 km	5 km	15 km	42 km	5 km	15 km	42 km
60°N–40°N	24.3	11.02	38.4	–0.74	0.45	–0.35	–0.90	–0.24	–0.76	–0.95	–0.31	–0.80
40°N–20°N	15.9	16.6	12.8	–0.76	–0.78	0.28	–0.57	–0.56	0.10	–0.75	–0.46	–0.55
20°N–0°	6.4	4.0	9.1	–0.10	–0.61	–0.03	–0.21	–0.42	0.32	–0.22	–0.30	0.45
0°–20°S	5.6	5.3	9.4	–0.16	0.07	–0.16	–0.62	0.08	0.47	–0.74	–0.08	0.45
20°S–40°S	15.4	15.6	19.7	–0.86	0.64	–0.38	–0.42	–0.56	–0.81	–0.37	–0.39	–0.80
40°S–60°S	18.2	11.0	34.0	–0.78	–0.15	–0.31	–0.76	–0.25	–0.85	–0.78	–0.53	–0.88

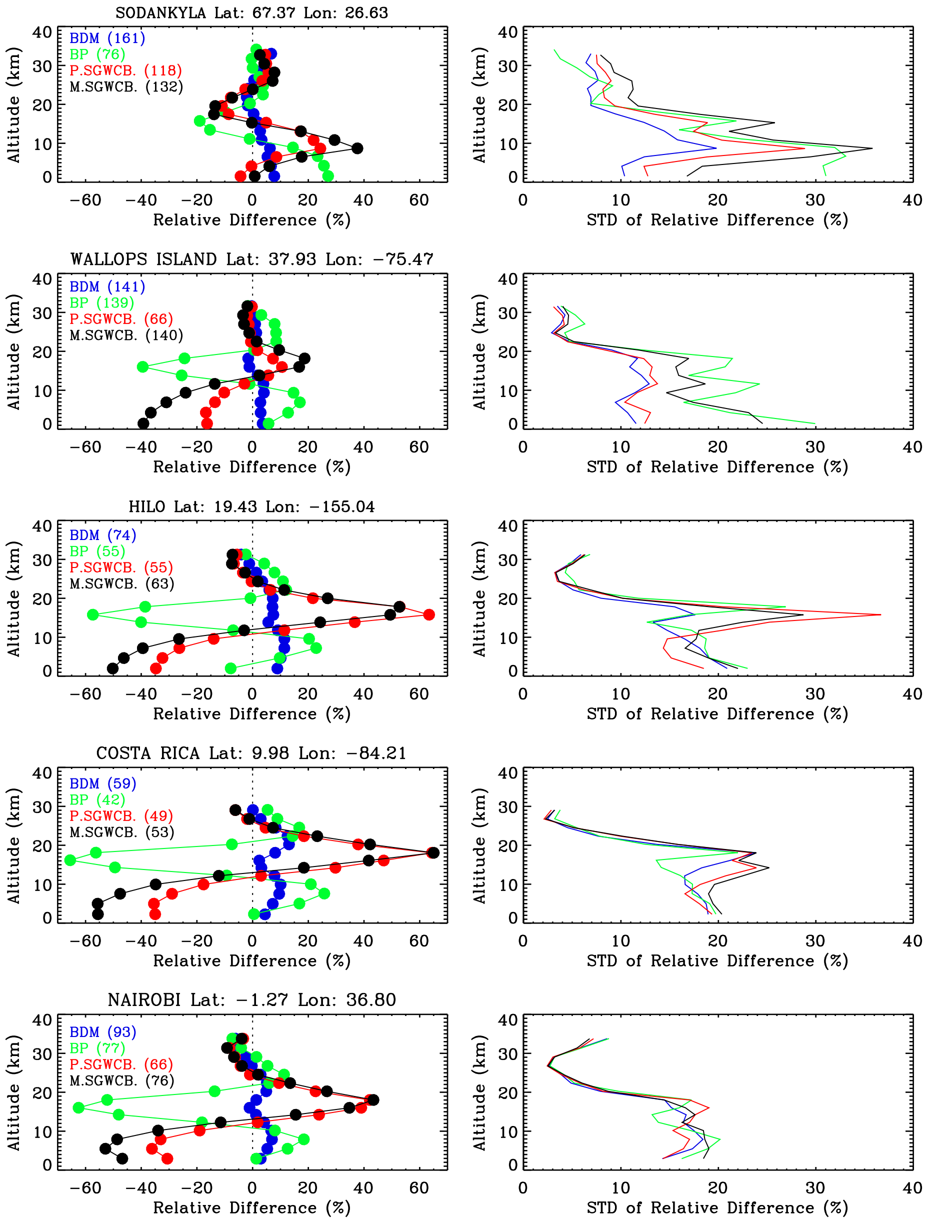

Fig. 11. Comparison of mean differences (left) and their standard deviations (right) between retrieved ozone profiles using different cross sections and ozonesonde observations. Ozonesonde observations are convolved with retrieval averaging kernels. The numbers in parenthesis indicate the number of collocated retrievals.

dataset being noisier. There are significant retrieval differences, especially below ~20 km, among the datasets. Relative to BDM retrievals, retrieved total ozone typically shows negative biases within 5 DU for BP and parameterized SGWCB data and negative biases of 5–10 DU for the original SGWCB data. The ozone differences are anti-correlated with the effective temperature, indicating they can well be explained by the temperature variation and different temperature dependence of the cross sections. The retrieved tropospheric O_3 columns generally have large negative biases of 5–10 DU and 5–20 DU for parameterized and original SGWCB, respectively. The retrieved ozone profiles with BP and SGWCB data on average show altitude-dependent oscillating differences (biases) of up to ±20–40% below ~20 km, with almost opposite bias patterns between the BP and SGWCB datasets. The ozone differences with respect to BDM retrievals at ~5 km for both BP and SGWCB datasets and at ~42 km for the SGWCB dataset show strong anti-correlation of 0.7–0.95 when the temperature variation is significant, confirming the linkage of ozone differences to different temperature dependences. To validate the retrievals using different O_3 cross sections, we compare them with collocated ozonesonde observations at five stations for 2004–2008. The BDM retrievals agree well with ozonesonde observations, typically within 10%. Similar to comparison with one orbit of BDM retrievals, both BP and SGWCB retrievals consistently show large altitude-dependent oscillating biases of up to $\pm$20–70% (depending on the station) below 20 km but with almost opposite bias patterns.

Based on the evaluation, we recommend the use of BDM cross sections for ozone profile retrievals from UV measurements and archiving this dataset in the HIgh-resolution TRANsmission molecular absorption (HITRAN) database. The better performance is likely due to better characterization of the temperature dependence in the Hartley and Huggins bands. The wavelength calibration and the precision of the SGWCB data are comparable to the BDM dataset. The large altitude-dependent oscillating biases in the retrieved ozone profiles (using either parameterized and original data) and the presence of large cross section residuals after the quadratic parameterization suggest this dataset might contain systematic biases at some temperatures. The BP data are noisier than both BDM and SGWCB data.

Acknowledgments

This study is supported by the NASA Aura Science Team Grant (NNX11AE95G) and the Smithsonian Institution. We thank the OMI science team for providing OMI level 1b data, and Aura AVDC, WOUDC, CMDL, and SHADOZ and their data originators for providing ozonesonde measurements. Finally, we acknowledge the two anonymous reviewers for providing constructive comments.

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