Impact of ozone cross-section choice on WFDOAS total ozone retrieval applied to GOME, SCIAMACHY, and GOME-2 (1995-present)

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Content

1 Introduction

The DOAS ozone retrieval applied to GOME, SCIAMACHY, and GOME-2 satellite spectral data has achieved a high level of maturity when compared to ground data, in particular, to Dobson and Brewer instruments (usually to within 1%). Currently, three DOAS type algorithms are available that have been successfully applied and they all show very good agreement among each other and with ground data (Bracher et al., 2005, Eskes et al., 2005, Coldewey-Egbers et al., 2005, Weber et al. 2005, van Roozendael et al. 2006, Balis et al., 2007). The GOME instrument (Burrows et al., 1999a), still in operation although with limited earth coverage since 2003, has been succeeded by SCIAMACHY which was launched aboard ENVISAT in 2002 (Bovensmann et al., 1999). GOME2 was launched in 2006 aboard METOP-2 (Callies et al., 2000). GOME, SCIAMACHY, and GOME-2 have nearly identical UV spectral channels so that the DOAS algorithms can be adapted to all instruments without large changes.

Some satellite DOAS algorithms are using ozone cross-sections from Burrows et al. (1999b) that were measured with the GOME flight model and are also known as the GOME FM O3 cross-sections. In the retrieval from Dobson and Brewer instruments the Bass-Paur cross-sections are standard (Bass and Paur, 1985, Paur and Bass, 1985, Staehelin et al., 2003). After proper adjustments to the spectral resolution of GOME, the Bass-Paur data yield total ozone that are 2% higher than that retrieved with the GOME FM spectra (van Roozendael et al., 2003). The idea of using GOME FM cross-sections in the GOME retrieval is the advantage that the instrument slit function does not need to be known. It is, therefore, obvious to use then SCIAMACHY FM cross-sections as reported by Bogumil et al. (2003) for SCIAMACHY and the GOME-2 FM cross-sections for GOME-2.

First results indicate that the use of the Bogumil data leads to a high bias on the order of +5% for SCIAMACHY with respect to the ground (Eskes et al., 2005) while the use of the convolved GOME FM cross-sections leads to an underestimation of about 2% (Eskes et al., 2005). In a preliminary analysis the use of the GOME FM cross-section in the WFDOAS SCIAMACHY retrieval showed good agreement with collocated GOME results (Bracher et al., 2005), but all studies used older calibration versions of the spectral level 1c data (V4.01 and V4.03). Bracher et al. (2005) also reported a high bias when using the SCIAMACHY cross-sections in the retrieval.

In this note we investigate how the choice of cross-sections impact the WFDOAS (Weighting Function Differential Optical Absorption Spectroscopy) total ozone retrieval (Coldewey-Egbers et al., 2005) for GOME, GOME-2, and SCIAMACHY.

As of March 2013 three new or updated ozone cross-section data are available: the high resolution cross-section data from Serdyuchenko et al. (2011, 2013, Gorshelev et al., 2013) and new versions of the SCIAMACHY FM (Chehade et al., 2013) and GOME-2 FM3 ozone cross-sections (Chehade et al., 2013a,b). These cross-sections are investigated in Section 8 (Serdyuchenko data) and briefly summarized in the Conclusion section (SCIA FM/GOME-2 FM3 data).

2 WFDOAS retrieval algorithm

The WFDOAS retrieval uses the Taylor expansion of the optical depth equation (Coldewey-Egbers et al., 2005) as follows:

$$\ln\frac{I_{obs}}{F_{obs}} = \boxed{\ln\left(\frac{I}{F}\right)_{mod}} + \boxed{+\frac{d\ln(I/F)}{d\mathrm{TOZ}}\Big|_{mod}}(\mathrm{TOZ}_{fit}-\mathrm{TOZ}_{clim}) + \boxed{+\frac{d\ln(I/F)}{d\mathrm{T}}\Big|_{mod}}(\mathrm{T}_{fit}-\mathrm{T}_{clim}) + \ldots + \mathrm{Pol} \quad , \qquad (1)$$

where I/F is the sun-normalized nadir radiance, *TOZ* the total ozone column and *T* the scalar temperature. The red squared boxes indicate radiation transfer quantities (*mod* for model), the first the reference optical depth for the model atmosphere, the two other red boxes are weighting functions for ozone and the scalar temperature. Other terms include the Ring effect, NO2 and BrO absorption and a polynomial, the last for broad-band effects like aerosol and scattering contributions (Coldewey-Egbers et al., 2005). The scalar parameter $\Delta T=T_{fit}-T_{clim}$ is the temperature shift applied to the temperature profile that is usually available along with the ozone profile climatology. The temperature sensitivity in the DOAS retrieval comes from the temperature dependence of the ozone cross-section in the Huggins band. From the temperature shift an effective ozone temperature can be derived. The ozone and temperature terms in Eq. 1 are shown in Fig. 1 for a selected GOME retrieval.

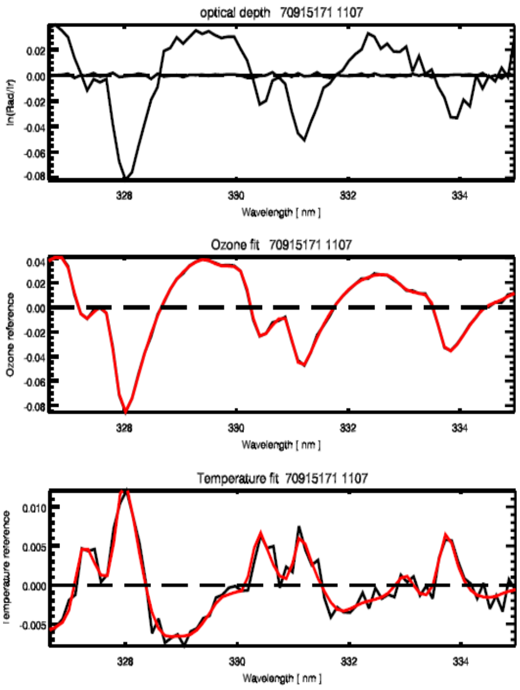

Figure 1. Ozone and temperature terms in the WFDOAS equation after a GOME retrieval. Top panel: GOME optical depth and fit residuals. Middle panel: fitted ozone term (red) and the same plus residual (black). Bottom panel: fitted temperature term (red) and the same plus residual (black) (Coldewey-Egbers et al., 2005).

In WFDOAS the fitting window is 326.6 nm-334.5 nm (GOME: 335 nm). As can be seen from Fig. 1, the ozone and temperature weighting function strongly anti-correlate. For the WFDOAS fitting windows the correlation is *r=-0.6* (Coldewey-Egbers et al., 2003). One has to keep this in mind when using retreived ozone temperature as a criterion for selecting the appropriate cross-section.

3 Available cross-sections

The following ozone cross-section data are considered here:

- GOME FM, spectral resolution: 0.17 nm @ 330 nm (Burrows et al., 1999b)
- SCIAMACHY FM, spectral resolution: 0.20 nm @ 330 nm (Bogumil et al., 2003)
- GOME2 FM3 Version 3, spectral resolution: 0.29 nm @ 330nm (Gür et al., 2006)
- Bass-Paur cross-section, spectral resolution: ~0.1 nm (Paur and Bass, 1985, Bass and Paur, 1985)
- Brion cross-section, spectral resolution: 0.01-0.02 nm (Daumont et al. 1992, Brion et al. 1993, 1998, Malicet et al. 1995)

The Brion et al. absorption cross-sections are absolute measurements at very high spectral resolution, which is convenient for convolution to satellite resolutions. All FM cross-sections are relative measurements which have been scaled to reference values at selected wavelengths (Burrows et al. 1999, Bogumil et al., 2003, Gür et al., 2006). The satellite FM spectra at about 220 K are shown in Figure 2.

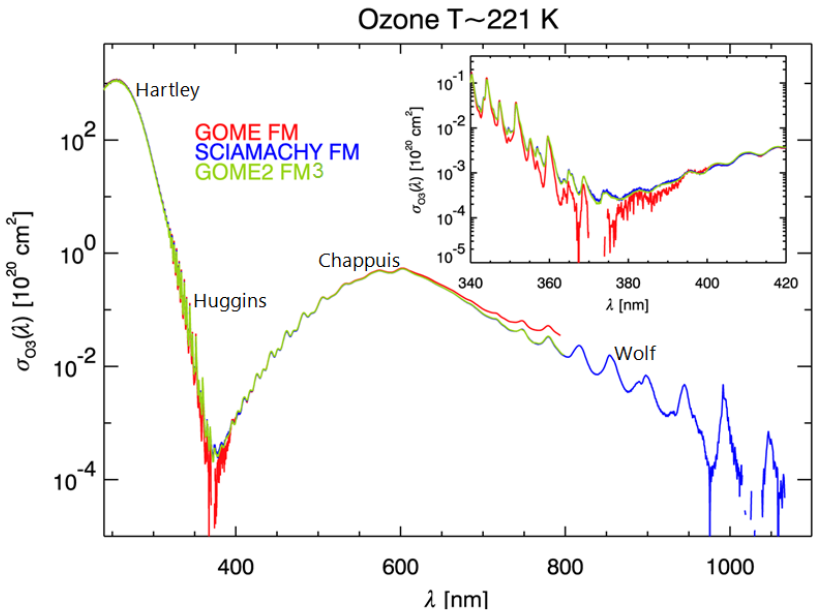

Figure 2: Flight model ozone absorption cross-sections from GOME, SCIAMACHY, and GOME2 at about 221 K.

All cross-sections considered here are available at four to five temperatures ranging from about 200 K to room temperature. In order to interpolate between temperatures a Bass-Paur parameterization is introduced as follows:

$$\sigma(\lambda, T) = a_o(\lambda)\left[1 + a_1(\lambda)\cdot T + a_2(\lambda)\cdot T^2\right] . \tag{2}$$

This parameterization is used in the radiation transfer code in combination with the temperature climatology to express the altitude dependent change of ozone absorption.

4 Diagnostics of ozone absorption cross-section

To look for differences between cross-sections it is useful to compare the different temperature coefficients in Eq. 2. Figure 3 shows the temperature coefficients for the satellite FM spectra. The coefficients agree well for SCIAMACHY and GOME, larger differences are observed for GOME2 FM spectra for the quadratic temperature coefficient. Small differences in spectral resolution (0.17 nm – 0.29 nm at 330 nm) explain some of the differences. There are jumps near 315 nm, which is due to the channel boundary between the two UV channels in GOME. Above 345 nm the ozone absorptions becomes weak and the data noisier. The spectral region between 335 nm and 380 nm is, nevertheless, an important spectral region, since most of the minor trace gases are retrieved here (BrO, OClO, H2CO, IO) and ozone remains a comparable and fairly strong absorber, which has to be accounted for. Figure 4 shows for comparison the temperature coefficients for Bass-Paur and Brion spectra. For better comparison with the satellite FM data they were convolved to GOME spectral resolution (~0.2 nm).

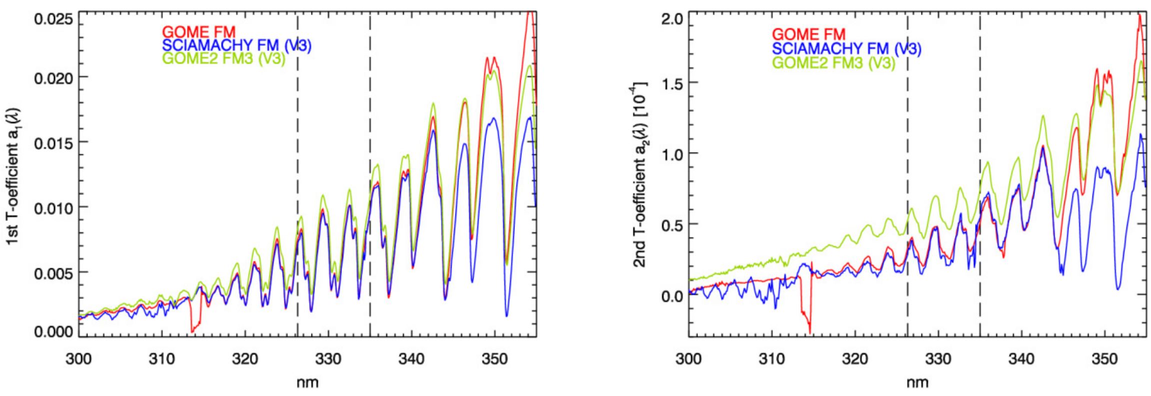

Figure 3: Temperature coefficients for satellite FM ozone absorption spectra. Left: $a_1(\lambda)$, right: $a_2(\lambda)$ (see Eq. 2). Vertical dashed lines indicate the DOAS fitting window.

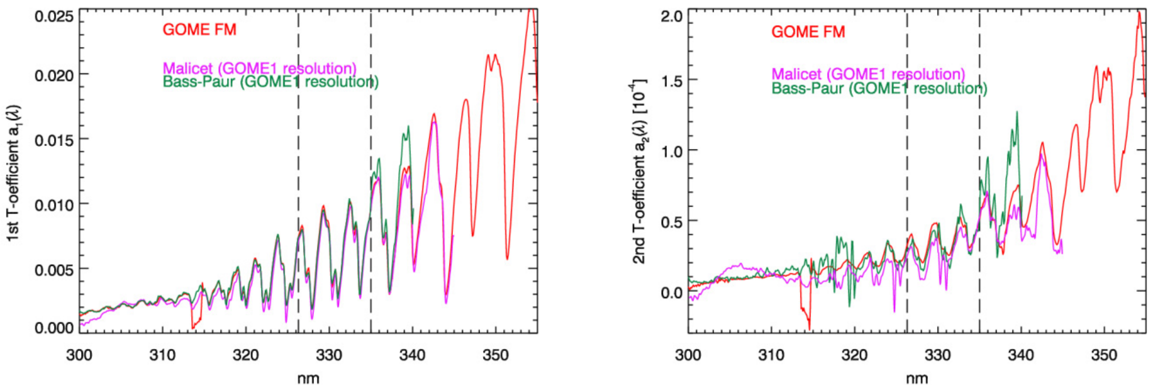

Figure 4: Same as Fig. 3, but for Brion (Malicet) and Bass-Paur. Left: $a_1(\lambda)$, right: $a_2(\lambda)$ (see Eq. 2). GOME FM data are shown for direct comparison.

It is evident that the Bass-Paur data appear to be noisier than Malicet, particularly in the 315 – 320 nm spectral range and above 335 nm. The quadratic temperature coefficient for Bass-Paur shows a clear jump right at 335 nm. This may indicate that the Bass-Paur data has been extended above 335 nm using, for instance, data from Vigroux (1953). The agreement between Bass-Paur and GOME FM is slightly better. One should keep in mind that selected Bass-Paur data at reference wavelengths were used to convert the relative GOME FM cross-section measurements to an absolute scale (Burrows et al., 1999b).

Another diagnosis of the quality of ozone cross-section data is the smoothness of the temperature behaviour of the actual cross-section data in comparison to Eq. 2. This is demonstrated in Fig. 5, where the temperature dependence from Eq. 2 is compared with measured cross-sections near an absorption minimum (328 nm) and maximum (330 nm). In general the differences of the measured cross-section and the smooth temperature behaviour of Eq. 2 are within 1%. At certain temperatures larger deviations are observed, for instance, for Bass-Paur data at 218 K, where differences reach 3% at 218 K. On the ACSO homepage (http://igacoo3.fmi.fi/ACSO/cross_sections.html) a different Bass-Paur data set is listed than is available from the MPI spectral database (http://www.atmosphere.mpg.de/enid/Spectra/Presentation_4n3.html). The MPI data base contains the original spectra from Bass-Paur (Paur and Bass, 1985) at five representative temperatures (203 K, 218 K, 228 K, 243 K, 273 K, 298 K). The Bass Paur data at the ACSO page appear to be spectra at some selected temperatures that were calculated from the polynomial (Eq. 2) fitted to the original data excluding the 218 K data.

It appears that the SCIAMACHY FM ozone cross-sections at absorption maximum is maller than GOME FM, although the spectral resolution is almost identical. This can explain in parts the bias seen in the SCIAMACHY total ozone using the Bogumil data. Lower values for ozone maxima means that the retrieved total ozone gets larger. The lowest temperature available for the Brion (Malicet) cross-sections is 218 K, so that extrapolation to lower temperatures close to polar stratosphere cloud (PSC) temperatures, typically seen inside the Antarctic vortex, leads to larger errors.

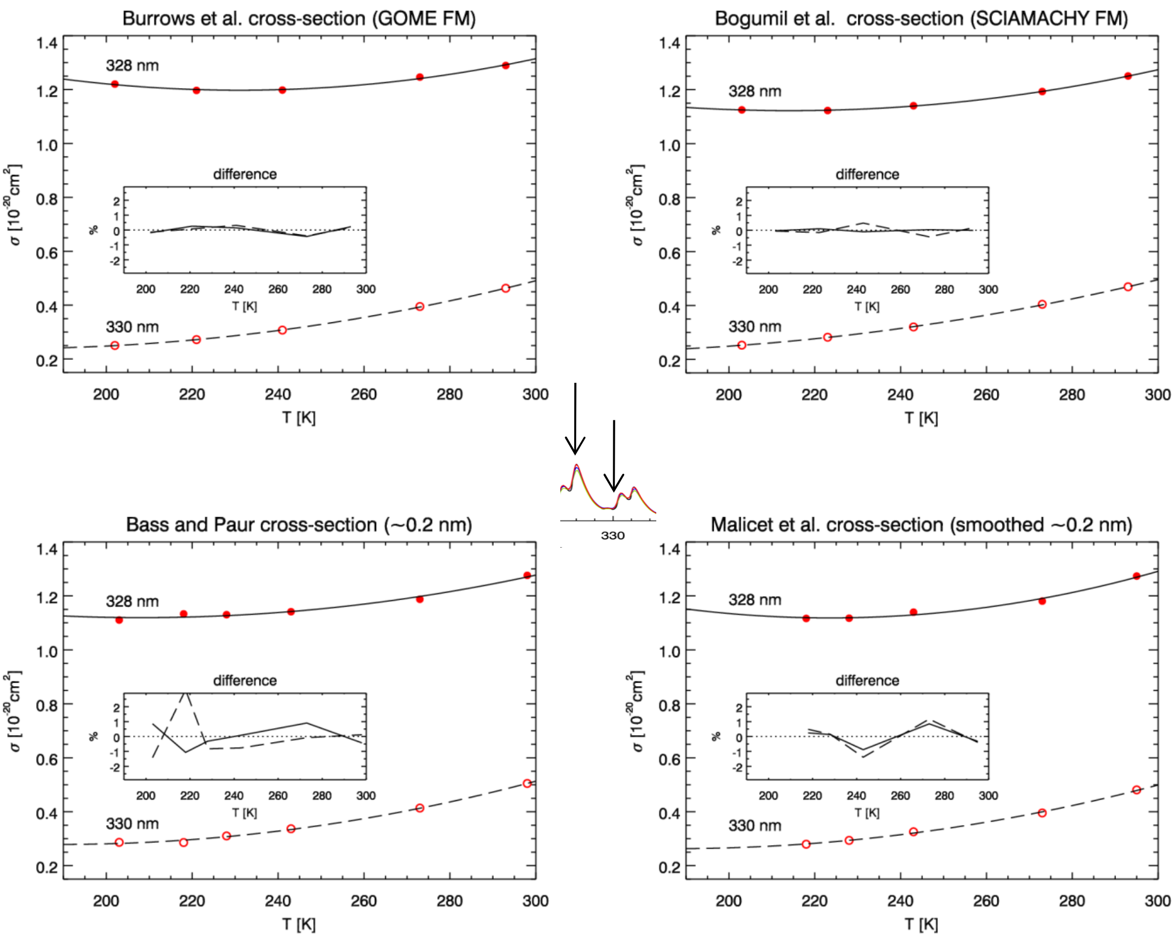

Figure 5: Measured ozone cross-sections (points) and temperature parameterisation (black lines) from Eq. 2. Differences between both are shown in the inlet.

5 Direct comparisons of cross-sections

In principle the cross-sections can be directly compared with each other if one accounts for the different spectral resolution. This can be done by

convolving the reference cross-section with an instrumental line shape (ILS, here approximated by a Gaussian) and then scaling to the lower resolution cross-section. This procedure is carried out by a non-linear least-square fit, where the fit parameters are the scaling factor, wavelength shift, Gaussian width of ILS, and a cubic polynomial (DOAS like fit).

For this fitting a Marquardt-Levenberg non-linear least squares estimation is applied to match the reference spectra $f_{ref}(\lambda)$ to the lower spectral resolution spectrum $f(\lambda)$ as follows:

$$f(\lambda) = a_o \cdot f_{ref}(\lambda - a_1) \otimes ILS(a_2, \lambda) + \sum_{i=0}^{3} a_{3+i} \cdot \lambda^i = \\ = a_o \cdot \int_{-\infty}^{\infty} f_{ref}(\lambda' - a_1) \cdot ILS(a_2, \lambda' - \lambda)\ d\lambda' + \sum_{i=0}^{3} a_{3+i} \cdot \lambda^i \tag{3}$$

where λ is the wavelength, a_i are fitting coefficients, and *ILS* the instrumental line shape here defined as a Gaussian with FWHM a_2, i.e.

$$ILS(a_2, \lambda' - \lambda) = N^{-1} \exp\left(-\ln 2 \frac{(\lambda' - \lambda)^2}{a_2^2} \right). \tag{4}$$

N is a normalization constant determined from the discretisation of the *ILS.* The reference spectrum is convolved with the ILS as indicated by the convolution sign $\otimes$. The use of a polynomial makes this a differential fitting (DOAS type fitting). a_o is interpreted as a scaling constant (like a slant column density in DOAS fitting) and a_1 represents a wavelength shift in the wavelength calibration.

In cases where the reference spectra have a finite and non-negligible slit width, the true FWHM is then approximated by the squared sum of the known slit width of reference data a_{ref} and the retrieved FWHM a_2 as follows

$$FWHM = \sqrt{a_{ref}^2 + a_2^2}\ . \tag{5}$$

Here we assume that the slit function is again a Gaussian and a consecutive convolution with two Gaussians is equivalent to square summing the two widths and applying one Gaussian convolution.

In the direct comparison of cross-sections, data with the Bass-Baur parameterization (Eq. 2) is used to match the temperatures. Table 1 summarises the results. A similar fitting can be applied to solar irradiance data from GOME, SCIAMACHY, and GOME-2 using high-resolution solar data from the Kitt Peak Observatory (Kurucz, 1995) as reference ("Fraunhofer fitting"). The FWHM retrieved from the solar fitting can be compared with the estimate from the cross-section fitting. The agreement is within the error bars very good, thus validating our approach for the direct cross-section comparisons. Both SCIAMACHY and GOME have symmetric ILS that is quite well represented by the Gaussian. In case of GOME-2 the ILS is asymmetric. In this case a two sided Voigt profile with different half widths was used (Van Roozendael et al., 2003). The parameters for the asymmetric ILS was retrieved from the solar fitting and kept fixed in the cross-section comparison since the wavelength shift correlates with the asymmetry of the ILS. Only a wavelength shift was here allowed.

Table 1. Direct comparisons of satellite FM spectra with Brion ozone cross-sections as reference in the 326.6 nm-334.5 nm fitting window.

Cross-sections	T[K]	Shift [nm]	Scaling [-]	Gaussian FWHM [nm]	Solar FWHM [nm]	Scaling wrt. GOME FM
Burrows et al. 1999 GOME FM	225 240	+0.017(2) +0.017(2)	1.027(2) 1.023(2)	0.158(5) 0.159(6)	0.165(8)	- -
Bogumil et al. 2003 SCIAMACHY FM	225 240	+0.008(2) +0.009(2)	0.970(3) 0.973(3)	0.222(6) 0.219(6)	0.202(13)	-5.6% -4.9%
Bass & Paur, 1985 (MPI database)	225 240	+0.020(3) +0.023(5)	1.015(3) 1.000(3)	0.079(10) 0.087(10)	-	+1.2% +2.3%
Bass & Paur, 1985 (ACSO web page)	225 240	+0.024(4) +0.021(4)	1.004(3) 1.004(3)	0.084(12) 0.078(11)	-	+2.3% +1.9%

The GOME FM cross-section requires a scaling of about -2 to -3% depending on temperature to match the Brion data. The SCIAMACHY FM needs to be scaled by about +5% to match GOME FM. A larger scaling is required for GOME2 FM3.

The wavelength shifts with respect to Brion data are positive for GOME and SCIAMACHY. The wavelength shifts for GOME-2 are negative and fairly large compared to SCIAMACHY and GOME (nearly 40% of the detector sampling interval). A proper wavelength shift is important to minimize the fit residual in the ozone retrieval. In order to compare the shifts from Table 1 to rms minimizing shifts in the DOAS ozone retrieval, the cross-sections were tested in the ozone retrieval using various shifts. As shown in Fig. 5 the ozone fit residuals are generally minimized at the shifts given in Table 1. One should note that total column changes are minus 2% per 0.01 nm shift in the cross-sections.

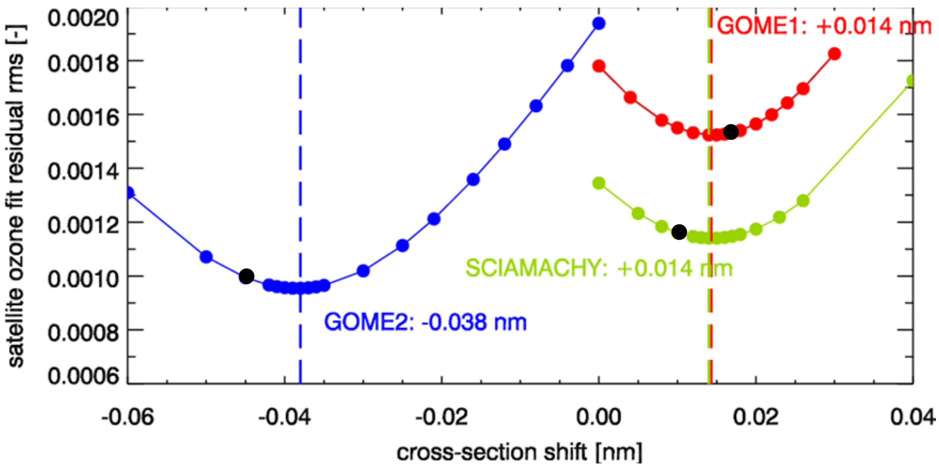

Fig. 6: Spectral fit residuals as a function of wavelength shifts in the respective FM ozone cross-sections. Black dots indicate the shifts as derived from the direct comparison of cross-sections with Brion data.

The good agreement in the shifts from the cross-section comparison and minimization of the ozone fit residuals prove that the Brion data have a very good wavelength stability near 330 nm. From Fig. 5 it is also demonstrated that the ozone fit residuals improved from GOME and SCIAMACHY to GOME-2. One of the reasons is that the GOME-2 data are clearly sampled more than twice per resolution element, while GOME is undersampled (1.5 detector pixel per resolution element), which adds noise.

The Bass-Paur data compare well with the Brion cross-sections in the DOAS windows (scaling differences are within 0.5%, Table 1). The spectral resolution of Bass-Paur seems however to be 0.1 nm FWHM, which is lower than cited in Orphal (2002, 2003). In the classical DOAS fitting window a wavelength shift of +0.023 nm is required to match Brion data and to minimize spectral residuals.

6 WFDOAS total ozone retrievals

As a first test, SCIAMACHY retrievals were carried out using different ozone cross-sections with the appropriate shifts from Tables 1 and 2. The spectral fit residuals are shown in Fig. 7.

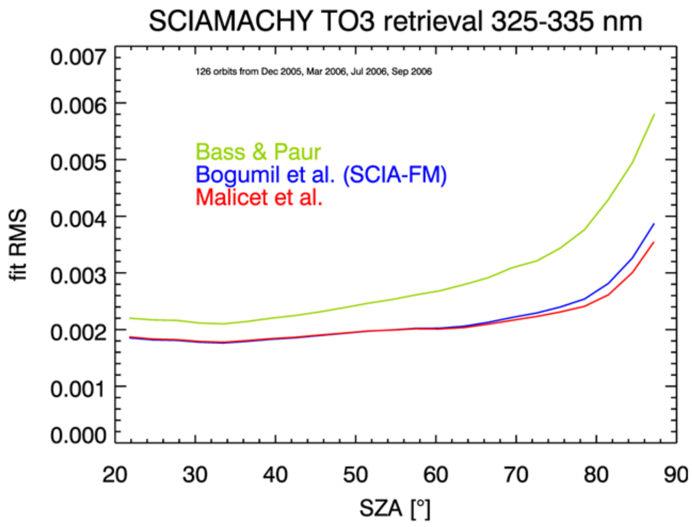

Figure 7: Spectral residuals as a function of solar zenith angle from SCIAMACHY WFDOAS retrievals using 126 satellite orbits during 2006.

The spectral fit residuals are lowest for the satellite FM (also for slit function adjusted GOME FM cross-sections not shown here). The use of Bass-Paur cross-section data leads to higher residuals, while Brion data are comparable and slightly better than SCIAMACHY FM at high SZA.

For one SCIAMACHY and GOME-2 orbit on October 1, 2008, different cross-sections have been used in the column retrieval. Results are summarised in Fig. 8. For both SCIAMACHY and GOME-2 the use of the Bass-Paur data leads to higher fit residuals similarly to that shown in Fig. 7. Using the Brion and Bass-Paur data (ACSO version) leads to a bias of about 2% to 2.5%, which is consistent with the scaling factor derived for the GOME FM cross-sections in Table 1. The scaled SCIAMACHY FM data leads to similar results than GOME FM data which is not unexpected. The spectral fit residual rms is about the same for Brion and the satellite FM cross-section data, in the case of GOME-2 Brion data are somewhat better. The ozone temperature, which is the ozone weighted temperature profile average changes for a few K when satellite FM data are replaced by Bass-Paur (ACSO version) and Brion data as shown in Fig. 8. The original Bass Bass Paur data parametrised by Eq. 2 including all available temperatures leads to some different temperature dependence and, thus, an ozone temperature dependent (latitude dependent) bias with respect to the other cross-sections.

Within 1% the changes in the ozone columns are consistent with the scaling derived from the direct comparisons of cross-sections done in the previous section.

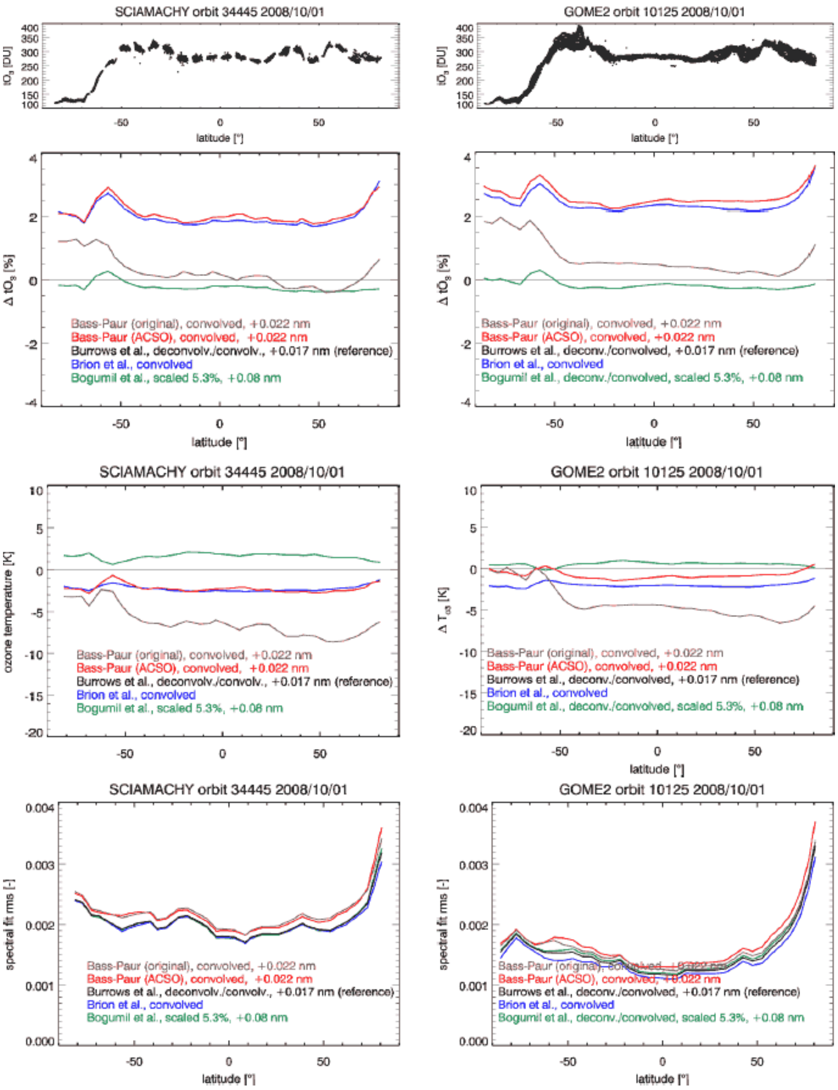

Figure 8. Use of different ozone cross-sections in ozone column WFDOAS retrieval applied to SCIAMACHY orbit 34445 (left column) and GOME-2 orbit 10125 (right column). Top row: Reference ozone columns calculated with resolution adjusted GOME FM cross-section. Second row: difference in O3 columns in percent. Third row: difference in retrieved ozone temperature in K. Third row: rms of fit residuals.

7 Merged GOME/SCIAMACHY/GOME-2 total ozone data set

For the multi-annual WFDOAS total ozone retrieval of GOME, SCIAMACHY, and GOME-2 following cross-section settings are used:

- GOME: Burrows et al. (1999b) cross-sections (GOME FM data), without scaling, shift: 0.17 nm
- SCIAMACHY: Bogumil et al. (2003) cross-sections (SCIAMACHY FM data), scaled by 5.3%, shift 0.09 nm
- GOME-2: Burrows et al. (1999) cross-sections, shift 0.017 nm, and convolved with GOME-2 slit function

These settings were derived from the direct comparisons of the cross-section data (Table 1). Figure 9 shows the global ozone from the three instruments since 2002. Also shown are adjusted SCIAMACHY and GOME-2 data records from comparisons to GOME during the period 2002-2008. Unadjusted SCIAMACHY data show a downward trend of about -4% per decade when compared to other satellite data and ground data (Lerot et al.

2009). The GOME-2 data record has a bias of about -1% with respect to GOME. Figure 10 shows how the merged GOME/SCIAMACHY/GOME-2 WFDOAS ozone data set compares to the SBUV/TOMS/OMI merged data set (Frith et al., 2004) and ground data (Fioletov et al., 2001). Agreement to the other data sets is very good.

With the cross-sections settings chosen for GOME, SCIAMACHY, and GOME-2 total ozone retrieval, differences are a few percent between instruments (Fig. 8). Some of the biases and drifts with time are due to calibration and instrumental artefacts. These errors are comparable to the differences seen when using different ozone cross-sections.

As of March 2013, GOME has ceased operation in early July 2011 and ESA lost communication with ENVISAT and SCIAMACHY in April 2012. GOME-2 aboard Metop-B has been launched in 2012. The WFDOAS merged datasets have been continued and are regularly used to show the evolution of global ozone (Weber et al., 2013).

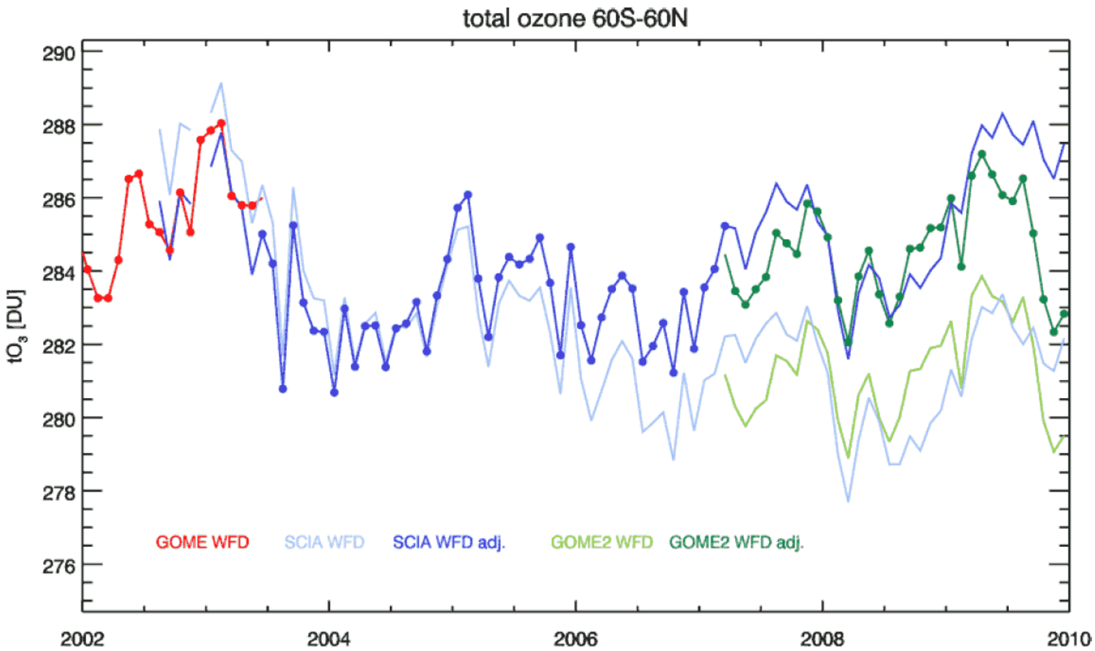

Fig. 9: The GOME, SCIAMACHY, and GOME-2 WFDOAS global ozone record. Also shown are the adjusted SCIAMACHY and GOME-2 data (dark blue and dark green) by comparison with GOME data during the period 2002-2008 (see http://www.iup.uni-bremen.de/gome/wfdoas/wfdoas_merged.html for details on the adjustments). The solid points indicate the data that become part of the GOME/SCIA/GOME-2 merged data record. The data have been deseasonalised and the annual mean of the seasonal fit added back.

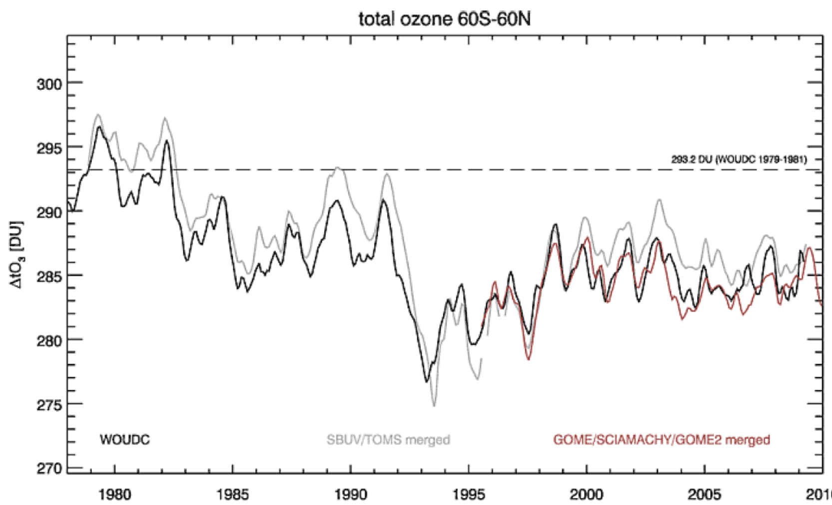

Figure 10. Global total ozone (60°S-60°N) from different data sets: WOUDC ground data (Fioletov et al., 2001), the SBUV/TOMS/OMI merged data set (Frith et al., 2004), and the merged GOME/SCIAMACHY/GOME-2 WFDOAS data set (http://www.iup.uni-bremen.de/gome/wfdoas/wfdoas_merged.html).

8 Impact of Serdyuchenko cross-section data (Update Nov. 2013)

In the following we extend our analysis to include the new Serdyuchenko et al. ozone cross-sections (Gorshelev et al. 2013, Serdyuchenko et al. 2011, 2013). Table 2 details the direct comparisons of the Bass-Paur (BP) and Brion data (BMD) with the Serdyuchenko cross-section data in the DOAS windows 325-335 nm. Each cross-section data has been fitted to a quadratic polynomial in temperature according to Eq. 2 and then compared at selected temperatures using Eq. 3. In order to match the differential absorption to Serdyuchenko et al. data a scaling of +1.4% to the Brion data has to be applied. The Bass Paur data requires a scaling of +1.0% and a wavelength shift of around 0.029 nm to match Serdyuchenko et al. data. A scaling of +1% and +1.4% of the BP and BMD cross-sections, respectively, to match Serdyuchenko data means that the use of Serdyuchenko et al. cross-sections in the DOAS total ozone retrieval will yield 1% and 1.4% higher total ozone compared to BP and BMD.

Table 2. Direct comparisons of Brion et al. and Bass Paur ozone absorption cross-sections to the new Serdyuchenko et al. cross-section data in the 325-335 nm window. Scaling factors and shift has to be applied in order to match the Serdyuchenko et al. data. Errors are given as 1σ

Cross-sections	Temperature [K]	Scaling factors [-]	Shift [nm]
Brion et al.	218	1.0128(10)	-
	227	1.0145(09)	-
	238	1.0142(12)	-
Bass Paur (ACSO)	218	1.0096(31)	0.0307(8)
	227	1.0115(29)	0.0292(7)
	238	1.0119(29)	0.0278(7)

Figure 11 shows a comparison of WFDOAS total ozone retrievals applied to one GOME-2 orbit using the different ozone cross-sections. The results can be summarized as follows:

- *Spectral fit residuals using BMD and Serdyuchenko data is lower than BP*
- *BP spectral fit residuals gets reduced if the BP are shifted by +0.029 nm, but remain higher than BMD and Serdyuchenko*
- *Serdyuchenko retrieved total ozone is on average ~1.5% higher than BMD and wavelength shifted Bass Paur*
- *BMD and wavelength shifted Bass Paur agree within 0.5%*
- *Without shifts in the BP cross-sections the total ozone gets about 5% higher*
- *Ozone results are as expected from the direct comparisons between cross-sections, see Tables 1 and 2*

Figure 12 shows the differences in the retrieved effective ozone temperature ($T_{O3,}$ ozone profile weighted temperature) from using the different ozone cross-sections. The differences in retrieved ozone temperatures between BMD, BP shifted by +0.029nm, and Serdyuchenko are within 4K. Above 60°S BMD shows a cold bias of about 4K (ozone hole condition) but is equal to BP temperatures for the remaining part of the orbit. There is a small negative bias in ozone retrieved with BMD under Antarctic ozone hole condition. Serdyuchenko is for most part of the orbit 4K higher than BP shifted and BMD. Without shifts BP leads to a +25 K bias in retrieved ozone temperatures. Sensitivities of ozone differences with respect to T_{O3} varies and are on the order of -1% to +1% DU per 20 K change between BMD, BP, and Serdyuchenko et al. cross-sections (bottom panel of Fig.12).

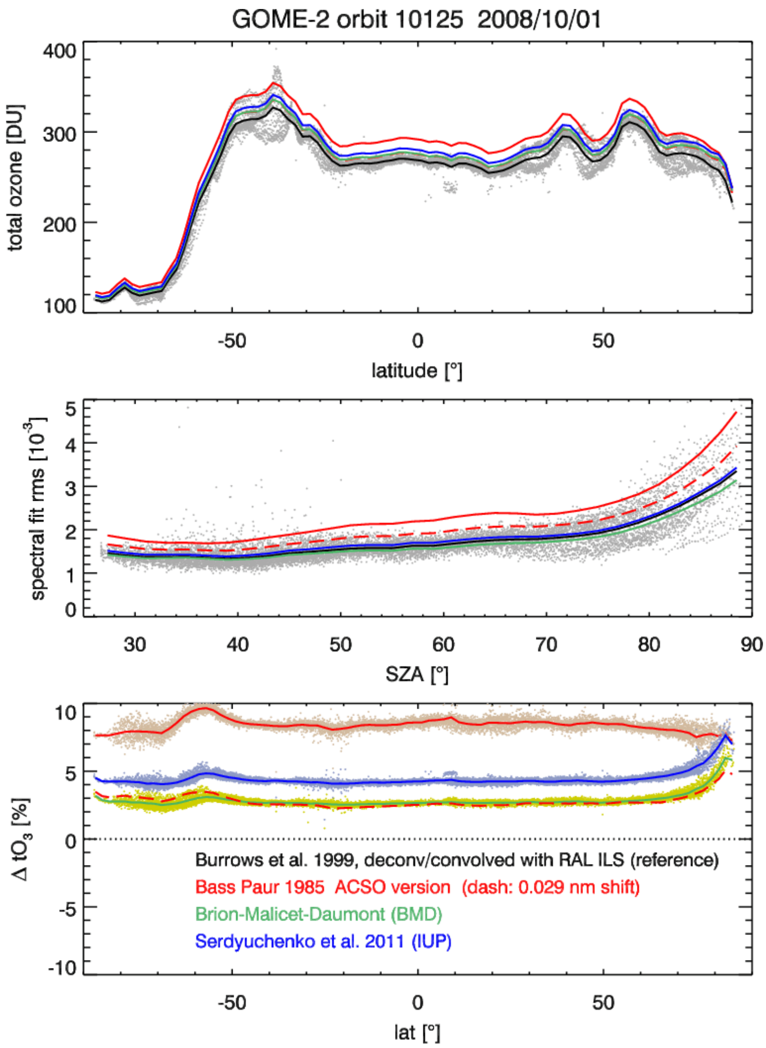

Figure 11: WFDOAS total ozone retrievals applied to one GOME-2 orbit (#10125 from October 1, 2008). Top panel: Retrieval using deconvolved Burrows et al. (1999) cross-sections with subsequent convolution with the GOME-2 slit function (RAL ILS). The individual data are shown as grey points while the black line shows the latitude binned results. The binned results from using the other cross-sections are shown as BP (red), BMD (green), and Serdyuchenko (blue). The middle panel shows the spectral fit residuals (RMS) from the use of the different cross-section data. The fit residuals of the retrieval with wavelength shifted BP data are also shown (dashed red line). Bottom panel: Differences in the retrieved total ozone with respect to the analysis using the Burrows et al. cross-section data.

9 Conclusion

Various cross-sections were tested in the WFDOAS retrievals in the classical ozone DOAS fitting window (325 – 335 nm). The proper use of satellite FM cross-sections in the ozone retrievals require proper wavelength shifts (see Table 1) in order to minimise the spectral fit residuals. The wavelength shifts derived from direct comparison to Brion cross-section data agree well with the shifts which minimise the spectral fit residuals. This proves that the Brion data are well wavelength calibrated near 330 nm. With proper wavelength shifts all satellite FM data and Brion cross-sections show comparable fit residuals, however, the Bass-Paur data appear noisier (see Fig. 4) than the others resulting in higher fit residuals (Figs. 7 and 11). This is also apparently the case in the 310-320 nm region (see Fig. 4) which is used in the Brewer and Dobson ground retrievals. It is clear from this study that better ozone cross-sections from more recent laboratory studies and with better signal-to-noise are now available that can supersede Bass-Paur cross-sections.

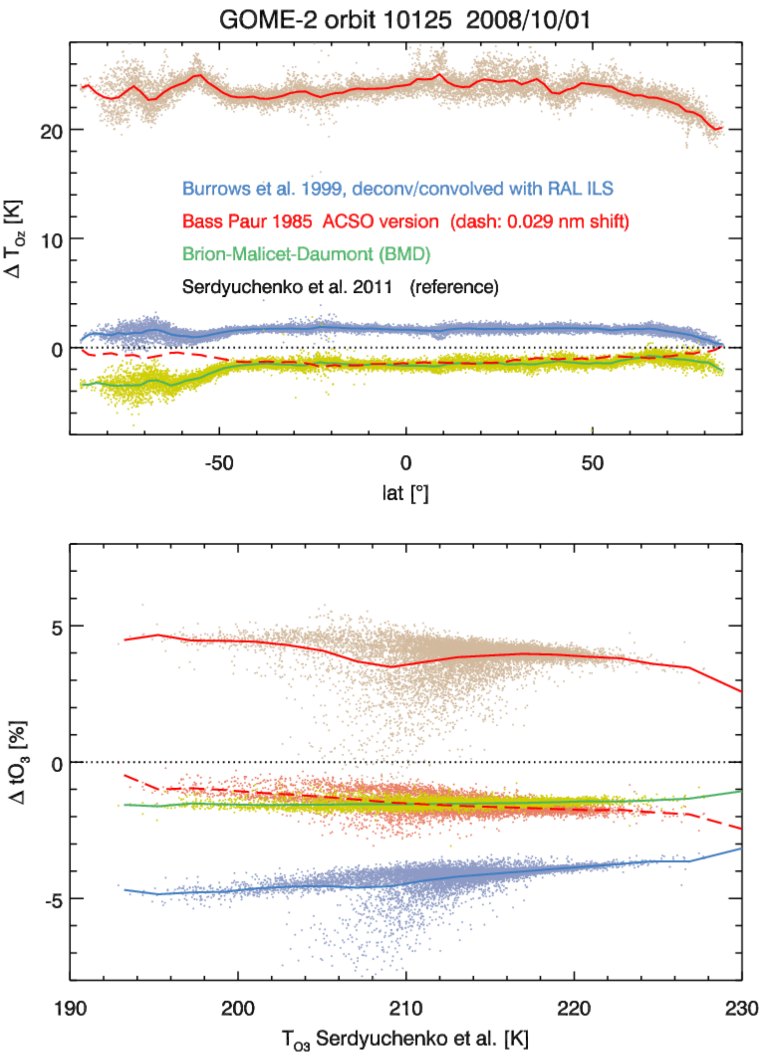

Figure 12: The same as Figure 11, but instead the difference in the retrieved effective ozone temperature to that from the Serdyuchenko retrieval is shown in the top panel. Bottom panel shows differences in retrieved total ozone as a function of effective ozone temperature.

The GOME FM and SCIAMACHY FM cross-sections are well suited for GOME and SCIAMACHY retrievals, respectively, but the SCIA FM (Bogumil et al.) needs to be scaled by 5% (see Table 1). The GOME2 FM3 Version 3 cross-sections (Gür et al., 2005) require a fairly large scaling of 8% and the wavelength shifts are quite large about 40% of the detector sampling of GOME-2. Fit residuals in the GOME-2 retrievals using these cross-sections are as low as for the convolved GOME FM cross-section data. *Very recently the GOME2 FM3 and SCIA FM cross-sections have been reanalysed and updated and their use in the total ozone retrievals now show differences below 1% with respect to the standard WFDOAS retrieval (Chehade et al., 2013a,b).*

The Brion ozone cross-sections (BMD) provide similar retrieval rms in the ozone fit as the satellite FM data, but a 2% scaling of GOME FM is required to match the Brion data. One disadvantage of the Brion cross-section is the lower limit of temperature range, which is high at 218 K and is far from the coldest temperatures inside the Antarctic ozone hole.

New laboratory measurements of ozone cross-sections were measured in our laboratory using a denser spacing of temperatures as shown in Fig. 5 for most of the current cross-section data and covering lower temperatures down to 193 K *(Gorshelev et al., 2013, Serdyuchenko et al., 2011, 2013)*. This should allow us to reduce uncertainties from the Bass-Paur temperature parameterisation (Eq. 2). It is important to keep in mind that the total ozone weighting function strongly anti-correlates with the temperature weighting function in the classical DOAS fitting window. *The Serdyuchenko cross-section data have been evaluated in the WFDOAS total ozone retrieval (Section 8). They deliver total ozone about 1.5% higher than BMD and wavelength shifted BP in agreement with direct comparisons between cross-section data in the classical DOAS window. There is an ozone temperature dependence in the total ozone differences that varies from -0.5%/10K to +0.5%/20K depending on which cross-section is used (shifted BP, BMD, and Serdyuchenko et al.). The spectral fit residuals from retrieving total ozone in the classical DOAS windows using Serdyuchenko et al. are similar to BMD and the other satellite FM cross-section data.*

Even with adjusted cross-section data or improved cross-section data, calibration uncertainties can cause biases between different instruments (see Fig. 8) that are of the same magnitude as differences from using different cross-section data.

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Appendix: Keywords

Figure A1: Word frequency map of this report providing the keywords. Created with the help of http://www.wordle.net (Courtesy: Jonathan Feinberg).