



# 1 Linearization of the effect of slit function changes 2 for improving OMI ozone profile retrievals

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8

## 9 **Abstract**

10 We introduce a method that reduces the spectral fit residuals caused by the slit function errors in an  
11 optimal estimation based spectral fitting process to improve ozone profile retrievals from the Ozone  
12 Monitoring Instrument (OMI) ultraviolet measurements (270-330 nm). Previously, a slit function was  
13 parameterized as a standard Gaussian by fitting the Full Width at Half Maximum (FWHM) of the slit  
14 function from climatological OMI solar irradiances. This cannot account for the temporal variation of slit  
15 function in irradiance, the intra-orbit slit function changes due to thermally-induced change and scene  
16 inhomogeneity, and potential differences in the slit functions of irradiance and radiance measurements. As  
17 a result, radiance simulation errors may be induced due to using the convolved reference spectra with  
18 incorrect slit functions. To better represent the shape of the slit functions, we implement a more generic  
19 super Gaussian slit function with two free parameters (slit width and shape factor); it becomes standard  
20 Gaussian when the shape factor is fixed to be 2. The effects of errors in slit function parameters on radiance  
21 spectra, referred as “Pseudo Absorbers (PAs)”, are linearized by convolving high-resolution cross sections  
22 or simulated radiances with the partial derivatives of the slit function with respect to the slit parameters.  
23 The PAs are included in the spectral fitting scaled by fitting coefficients that are iteratively adjusted as  
24 elements of the state vector along with ozone and other fitting parameters. The fit coefficients vary with  
25 cross-track and along-track pixels and show sensitivity to heterogeneous scenes. The total PA spectrum is  
26 quite similar in the Hartley band below 310 nm for both standard and super Gaussians, but is more distinctly  
27 structured in the Huggins band above 310 nm with the use of super Gaussian slit functions. Finally, we  
28 demonstrate that some spikes of fitting residuals are slightly smoothed by accounting for the slit function



29 errors. Comparisons with ozonesondes demonstrate substantial improvements with the use of PAs for both  
30 standard and super Gaussians, especially for reducing the systematic biases in the tropics and mid-latitudes  
31 and reducing the standard deviations at high-latitudes. Including PAs also makes the retrievals consistent  
32 between standard and super Gaussians. This study corroborates the slit function differences between  
33 radiance and irradiance demonstrating that it is important to account for such differences in the ozone profile  
34 retrievals.

35

## 36 1. Introduction

37 The fitting of the measured spectrum to the simulated spectrum is the most basic concept for the analysis  
38 of the Earth's atmospheric constituents from satellite measurements. Therefore, the accurate calibration and  
39 simulation of measurements are essential for the successful retrieval of atmospheric constituents. The  
40 knowledge of the instrumental spectral response function (ISRF) or slit function could affect the accuracies  
41 of both calibration and simulation, as it is required for the convolution of a high-resolution reference  
42 spectrum onto instrument's spectral resolution in the wavelength calibration and for the convolution of  
43 high-resolution absorption cross section spectra or simulated radiance spectrum in the calculation of  
44 radiance at instrumental resolution. Compared to other trace gases, the retrieval of ozone profiles could be  
45 more susceptible to the accuracy of ISRFs due to the large spectral range, where the radiance spans a few  
46 orders of magnitude and to the fact that the spectral fingerprint for the tropospheric ozone is primarily  
47 provided by narrow and weak absorption features of the temperature-dependent Huggins bands (320-360  
48 nm). Therefore, the efforts of characterizing and verifying the ISRFs have preceded the analyses of ozone  
49 profiles from the satellite/aircraft measurements (Liu et al., 2005, 2010; Cai et al., 2012; Liu et al., 2015;  
50 Sun et al. 2017; Bak et al., 2017).

51 For space-borne instruments, ISRFs are typically characterized as a function of the detector dimensions  
52 using a tunable laser source prior to the launch (Dirksen et al., 2006; Liu et al., 2015; van Hees et al., 2018).  
53 However, the preflight measured ISRFs could be inconsistent with those after launch due to the orbital  
54 movement and the instrument temperature change (Beirle et al., 2017; Sun et al., 2017). Therefore, the post-  
55 launch ISRFs have been typically parameterized through a cross-correlation of the measured solar  
56 irradiance to a high-resolution solar spectrum (Caspar and Chance, 1997), assuming Gaussian-like shapes.  
57 The direct retrieval of the ISRFs from radiances has not typically been done due to the complication of  
58 taking the atmospheric trace gas absorption and Ring effect into account in the cross-correlation procedure  
59 and the slow-down of the fitting process. However, slit function differences between radiance and  
60 irradiance could exist due to scene heterogeneity, differences in stray light between radiance and radiance,



61 intra-orbit instrumental changes, and the instrument temperature change (Beirle et al., 2017; Sun et al.,  
62 2017). In addition, using temporally invariant slit functions derived from climatological solar spectra in the  
63 retrievals could cause the long-term trend errors if instrument degradation occurs. Therefore, there is room  
64 for improving our trace gas retrievals by accounting for the effects of the different ISRFs between radiance  
65 and irradiance on the spectral fitting and on the pixel-to-pixel basis. The “Pseudo Absorber (PA)” is a  
66 common concept in spectral fitting to account for the effect of the physical phenomena that is difficult or  
67 computationally demanding to be simulated in the radiative transfer calculation, like spectral misalignments  
68 (shift and stretch) between radiance and irradiance, Ring effect, spectral undersampling, and additive stray-  
69 light offsets. The pseudo absorption spectrum can be derived from a finite-different scheme (e.g. Azam and  
70 Richter, 2015) or a linearization scheme via a Taylor expansion (e.g. Beirle et al., 2013; 2017); the latter  
71 approach is more efficient than the former one, but less accurate because only the first term of the Taylor  
72 series is typically taken into account for simplicity. Beirle et al. (2013) introduced a linearization scheme  
73 to account for spectral misalignments between radiance and irradiance and then included them as a pseudo-  
74 absorber in DOAS-based NO<sub>2</sub> and BrO fittings. Similarly, Beirle et al. (2017) linearized the effect of the  
75 change of the ISRF parameterized as a super Gaussian on GOME-2 solar irradiance spectra to characterize  
76 the slit function change over time and wavelength. Sun et al. (2017) derived on-orbit slit functions from  
77 solar irradiance spectra measured by the Ozone Monitoring Instrument (OMI) (Leveld et al., 2006) assuming  
78 standard Gaussian, super Gaussian, and preflight ISRFs with adjusted widths. The derived on-orbit slit  
79 functions, showing significant cross-track dependence that cannot be represented by preflight ISRFs,  
80 substantially improve the retrievals by the Smithsonian Astrophysical Observatory (SAO) ozone profile  
81 algorithm. However, it is not fully understood why the use of super Gaussian or stretched preflight functions,  
82 which are supposed to better model the OMI spectra as indicated by smaller mean fitting residuals, does  
83 not improve the retrievals over the use of standard Gaussian especially in the standard deviations of the  
84 differences with relative to ozonesonde observations. This study suggested that the slit functions derived  
85 from solar spectra might not fully represent those in radiance spectra.

86 As such, the objective of this paper is to implement the slit function linearization proposed by Beirle  
87 et al. (2017) into the optimal estimation based spectral fitting of the SAO ozone profile algorithm. We  
88 further improve the slit function parameterization by accounting for the differences between radiance and  
89 irradiance slit functions on a pixel-by-pixel basis, and ultimately to improve OMI ozone profile retrievals.  
90 This paper is organized as follows: after a mathematical description of the linearization of slit function  
91 changes using the generic super Gaussian function, we introduce how to apply them practically in an  
92 optimal estimation based spectral fit procedure (Section 2). This linearization scheme is differently  
93 implemented, depending on the simulation scheme of measured spectra using high resolution radiances or  
94 effective cross section data, respectively. Section 3 characterizes the derived pseudo absorber spectra, along



95 with the evaluations of ozone profile retrievals using independent ozonesonde observations as a reference  
96 dataset. Finally, the summary of this study is given in Section 4.

97 **2. Method**

98 **2.1 Super Gaussian linearization**

99 The slit function parameterization and linearization are briefly summarized from Beirle et al. (2017),  
100 focusing on what we need to derive the pseudo absorbers in the terms of the optimal estimation based fitting  
101 process. The slit function can be parameterized with the slit width  $w$ , and shape factor  $k$  assuming the  
102 super Gaussian,  $S$  as:

103

$$S(\Delta\lambda) = A(w, k) \times \exp\left[-\left|\frac{\Delta\lambda}{w}\right|^k\right], \quad (1)$$

104 where  $A(w, k)$  is  $\frac{k}{2\sigma_g \Gamma\left(\frac{1}{k}\right)}$  with  $\Gamma$  representing the gamma function. This equation allows many forms of  
105 distributions by varying  $k$ : the top-peaked function ( $k < 2$ ), the standard Gaussian function ( $k = 2$ ), and the  
106 flat-topped function ( $k > 2$ ).  $w$  is converted to the Full Width at Half Maximum (FWHM) via the relationship  
107 of  $\text{FWHM} = 2\sqrt{\ln 2} w$ . We investigate the impact of including one more slit parameter  $k$  on the OMI ISRF  
108 fit results over the standard Gaussian using OMI daily solar measurements. As an example, time-series  
109 (2005-2015) of the fitted slit width and shape factor in 310-330 nm are displayed in Figure 1.a. The FWHM  
110 and shape factor of the super Gaussian function is on average 0.44 nm and 2.9, respectively, while the  
111 FWHM of the standard Gaussian is 0.395 nm. The degradation of the OMI slit functions became relatively  
112 visible after 2011. The high wavelength stability (0.003 nm) is seen in Figure 1b, verifying that better  
113 calibration stability is performed with super Gaussian slit functions as abnormal deviations of wavelength  
114 shifts are derived with standard Gaussian slit functions.

115 The effect of changing the slit parameters  $p$  on the slit function can be linearized by the first-order  
116 Taylor expansion approximation around  $S_0 = S(p_0)$ :

117

$$\Delta S = S - S_0 \approx \Delta p \frac{\partial S}{\partial p}, \quad (2)$$

118 and thus the effect of changes of  $S$  on the convolved high-resolution spectrum can be parameterized as

119

$$\Delta I = I - I_0 = S \otimes I_h - S_0 \otimes I_h = \Delta S \otimes I_h, \quad (3)$$

120 where the convolved spectrum is  $I = S \otimes I_h$ . Consequently, the partial derivatives of  $I$  with respect to slit  
121 parameters,  $p$  are defined as



122 
$$\frac{\partial I}{\partial p} = \frac{\partial S}{\partial p} \otimes I_h. \quad (4)$$

123 In Beirle et al. (2017),  $\frac{\partial I}{\partial p}$  refers to  $J_p$  as “resolution correction spectra (RCS)”. In Figure 2, we present  
124 an example of  $J_p$  over the typical ozone profile fit range (270-330 nm) through the convolution of high-  
125 resolution ozone cross sections ( $\delta_h$ ) with the derivatives of the super Gaussian ( $\frac{\partial S}{\partial p}$ ). The baseline  $S_0$  is  
126 defined with  $w=0.26$  nm and  $k=2.6$ , which are averaged parameters from climatological OMI solar  
127 irradiance spectra in the UV2 band (310-330 nm). Note that this  $w$  value corresponds to a FWHM of 0.45  
128 nm. The change of the assumed OMI slit function causes a highly structured spectral response over the  
129 whole fitting window. However, the relative magnitude of the responses with respect to both slit parameters  
130 is more distinct in the Huggins band (>310 nm) where narrow absorption features are observed as shown  
131 in Figure 2.a. An anti-correlation (-0.92) is found between  $\frac{\partial \ln \delta}{\partial w}$  and  $\frac{\partial \ln \delta}{\partial k}$  while the response of the unit  
132 change of the slit width to the convolved spectrum is dominant against that of the shape factor.

133

## 134 **2.2 Implementation of the slit function linearization in the SAO ozone profile algorithm**

135  
136 We implement the slit function linearization in the SAO ozone profile algorithm (Liu et al. 2010), which  
137 is routinely being performed to produce the OMI PROFOZ product  
138 (<https://avdc.gsfc.nasa.gov/index.php?site=1389025893&id=74>). Two spectral windows (i.e., 270-309 nm  
139 in the UV1 band and 312-330 nm in the UV2 band) are employed to retrieve ozone profiles from OMI BUV  
140 measurements. To match the different spatial resolutions between UV1 and UV2 bands, every two cross-  
141 track pixels are averaged for UV2 band, resulting into 30 positions with the spatial resolution of 48 km  
142 (across-track)  $\times$  13 km (along-track) at nadir position. The non-linear optimal estimation based fitting is  
143 iterated toward minimizing the fitting residuals between measured and simulated radiances and  
144 simultaneously between a priori and estimated ozone values. A priori ozone information is taken from a  
145 tropopause-based (TB) ozone profile climatology (Bak et al., 2013). The Vector Linearized Discrete  
146 Ordinate Radiative Transfer model (VLIDORT) (Spurr, 2008) is used to simulate the radiances and their  
147 derivatives with respect to geophysical parameters. The radiance calculation is made for the Rayleigh  
148 atmosphere, where the incoming sunlight is simply absorbed by ozone and other trace gases, scattered by  
149 air molecules, and reflected by surfaces/clouds assumed as a Lambertian surface. Besides these physical  
150 phenomena, the others are treated as PAs to the spectral response such as Ring effect, additive offset, and  
151 spectral shifts due to misalignments of radiance relative to irradiance and ozone cross sections. In the SAO  
152 algorithm, these PAs are derived using the finite differences of the radiances with and without perturbation



153 to a phenomenon, except for the Ring spectrum that is calculated using a first-order single scattering  
154 rotational Raman scattering model (Sioris and Evans, 2000).

155 In this paper, we introduce new PAs to account for the radiance simulation errors caused by the slit  
156 function errors. The OMI ISRFs have been parameterized as a standard Gaussian from climatological OMI  
157 solar irradiances for each UV1 and UV2 band and thereby these PAs could take into account the spectral  
158 fitting responses caused by temporal variations of the slit function. This ozone fitting procedure uses ISRFs  
159 to convolve high resolution absorption spectra, taken from Brion et al. (1993) for ozone absorption cross  
160 section and Wilmouth et al. (1999) for BrO absorption cross section. Our algorithm has implemented two  
161 different convolution processes, i.e. the effective cross section approach in Liu et al (2010) and the high-  
162 resolution convolution approach described in Kim et al. (2013), respectively and thereby this paper also  
163 introduces how to derive the derivatives of the OMI radiances with respect to ISRF changes in these two  
164 approaches. Although the latter is the current approach, we also implement and present the linearization  
165 with the first approach, which is typically used for other trace gas retrieval algorithms.

166 In Liu et al (2010), VLIDORT simulates the radiances at OMI spectral grids ( $\lambda_{\text{omi}}$ ) using effective cross  
167 sections that are produced by convolving high-resolution cross sections with the OMI ISRF. Therefore, we  
168 apply a similar convolution process of matching the high-resolution cross section spectrum with OMI  
169 spectrum to derive the partial derivative of  $\sigma_x$  with respect to slit parameter, p as follows:

$$170 \quad \frac{\partial \sigma_x}{\partial p} = \frac{\partial S}{\partial p} \otimes \sigma_{x,h}, \quad (5)$$

171 where  $\sigma_{x,h}$  is a high-resolution absorption spectrum for ozone and BrO, respectively. Due to the dominant  
172 ozone absorption over the BrO absorption, the derivative of BrO cross section with respect to p is neglected  
173 here. This partial derivative of ozone is then converted to the partial derivative of radiance through the chain  
174 rule with the analytical ozone weighting function ( $\frac{d\ln I}{dO_3}$ ), calculated from VLIDORT, as follows:

175

$$176 \quad \frac{\partial \ln I}{\partial p} = \frac{\partial \ln I}{\partial O_3} \frac{\partial \sigma}{\partial p} \frac{O_3}{\sigma}. \quad (6)$$

177 This simulation process is hereafter referred to as “effective resolution cross section (ER) simulation”.

178 As described in Kim et al. (2013), the radiative transfer calculation in the SAO ozone profile algorithm  
179 has been performed using high-resolution extinction spectra at the optimized sampling intervals for  
180 resolving the ozone absorption features, which are a 1.0 nm below 300 nm and 0.4 nm above 300 nm. These  
181 sampling intervals are coarser than actual OMI sampling grids with approximately half the number of  
182 wavelengths. The coarser sampled simulated radiances are then interpolated to a fine grid of 0.05 nm  
183 assisted by the weighting functions with respect to absorption and Rayleigh optical depth:



184  $I(\lambda_h) = I(\lambda_c) + \frac{\partial I(\lambda_c)}{\partial \Delta_l^{gas}} \left( \Delta_l^{gas}(\lambda_h) - \Delta_l^{gas}(\lambda_c) \right) + \frac{\partial I(\lambda_c)}{\partial \Delta_l^{ray}} \left( \Delta_l^{ray}(\lambda_h) - \Delta_l^{ray}(\lambda_c) \right), \quad (7)$

185 where  $\Delta_l^{gas}$  and  $\Delta_l^{ray}$  are the optical thickness (the product of cross section and layer column density) at  
 186 each layer for trace gas absorption and Rayleigh scattering, respectively. The convolution is then applied  
 187 to these simulated high-resolution radiances,  $I(\lambda_h)$  with assumed slit functions and derivatives, respectively,  
 188 and thereby  $I(\lambda_{omi})$  and  $\frac{\partial \ln I}{\partial p}$  is calculated. This simulation process is hereafter referred to as “high-  
 189 resolution cross section (HR) simulation”. The ER simulation is more commonly implemented in the trace  
 190 gas retrievals in the UV and visible, but the HR simulation allows for more accurate fitting residuals to,  
 191 better than 0.1 % (Kim et al., 2013) as well as shorter computation time.  $\frac{\partial \ln I}{\partial p}$  is scaled by the fitting  
 192 coefficients,  $\Delta p$ , to account for the actual size of the spectral structures caused by the slit function  
 193 differences between radiance and irradiance spectra. The total “pseudo absorber (PA)” for the Super  
 194 Gaussian slit function linearization is expressed as:

195  $PA = \frac{\partial \ln I}{\partial k} \Delta k + \frac{\partial \ln I}{\partial w} \Delta w. \quad (8)$

196 In the form of the logarithm of normalized radiances, PA is physically related to the optical depth change  
 197  $\Delta \tau$ . Figure 3 compares the partial derivatives of radiances to slit parameters in HR and ER simulations.  
 198 Little difference is found even though convolution error for ozone cross sections is only accounted for in  
 199 the ER simulation due to the overwhelming impact of ozone cross section convolution errors over other  
 200 cross section data.

201 Furthermore, this linearization process can be formulated with n-order polynomial fitting parameters  
 202  $(\Delta p_i)$  to account for the wavelength-dependent change of the slit parameters around a central wavelength  $\bar{\lambda}$   
 203 and consequently, the total PA is expressed as

204  $PA = \frac{\partial \ln I}{\partial k} \sum_{i=1}^n \Delta k_i \cdot (\lambda - \bar{\lambda})^{n-1} + \frac{\partial \ln I}{\partial w} \sum_{i=1}^n \Delta w_i \cdot (\lambda - \bar{\lambda})^{n-1}. \quad (9)$

205

### 206 3. Results and Discussion

207 We characterize the effect of including the PA  $(\frac{\partial \ln I}{\partial p} \cdot \Delta p)$  on ozone profile retrievals using both Super  
 208 Gaussian and standard Gaussian slit functions. Hereafter, the correction spectrum  $(\frac{\partial \ln I}{\partial p})$  is derived using the  
 209 HR simulation. The PA coefficient  $(\Delta p_i)$  (one for each channel and for each order) is included as part of  
 210 the state vector to be iteratively and simultaneously retrieved with ozone. The a priori value is set to be zero



211 for all fitting coefficients, while the a priori error is set to be 0.1, empirically. We should note that the  
212 empirical “soft calibration” is applied to OMI radiances before the spectral fitting, in order to eliminate the  
213 wavelength and cross-track dependent systematic biases, due to the interference of the PA coefficients with  
214 systematic measurement errors during the fitting process.

215 **3.1 Characterization of the pseudo absorbers in ozone fitting procedure**

216 Figure 4 displays how the zero-order PA coefficients ( $\Delta p$ ) vary within one orbit when slit functions are  
217 assumed as standard and Super Gaussians, respectively, along with variation of cloud fraction, surface  
218 albedo, and cloud pressure from the retrievals. These fitting coefficients physically represent the difference  
219 of slit parameters between radiance and irradiance in this implementation. Therefore, we normalize them  
220 with the slit parameters derived from OMI solar irradiances for a better interpretation. Cross-track  
221 dependent features are shown in slit width. The relative change of the slit width is more distinct in the UV1  
222 band than in the UV2 band, whereas the change of the shape factor is more distinct in the UV2 band. The  
223 UV2 slit widths increase typically within 5 % over the given spatial domain. However, the UV1 slit widths  
224 increase from 10 % at most pixels up to 50 % at off-nadir positions in the high latitudes, which might be  
225 caused by stray light differences between radiance and irradiance and intra-orbit instrumental changes. An  
226 abnormal change of the UV1 slit parameters due to the scene heterogeneity is detected at the along-track  
227 scan positions of  $\sim 300$  and 900, respectively, where upper-level clouds are present. The UV2 shape factor  
228 changes show a coherent sensitivity to bright surfaces under clear-sky condition over the northern high  
229 latitudes. Fitting coefficients for the standard Gaussian show a quite similar spatial variation for the UV1  
230 slit width (correlation =  $\sim 0.98$ ), but an anti-correlation of  $\sim -0.62$  for the UV2 slit width compared to those  
231 for Super Gaussian due to the interference between shape factor and slit width.

232 Examples of the total PAs (eq. 9) are illustrated in Figure 5 when (a) zero and (b) first-order polynomial  
233 are fitted, respectively. The UV1 total PA spectrum, regardless of which Gaussian is assumed as slit  
234 function, is very similar because the spectral structure caused by the slit width change is dominant. It implies  
235 that OMI ISRFs in the UV1 band are similar to the standard Gaussian, for both radiance and irradiance  
236 measurements, consistent with the pre-launch characterization (Dirksen et al., 2006). However, in the UV2  
237 band, the PA is mostly contributed from the shape factor change in the case of super Gaussian, and the total  
238 PA spectrum is more noticeable for super Gaussian. Our results indicate that the PA for the shape factor  
239 change is required to adjust the spectral structures due to the differences in the slit functions between  
240 radiance and irradiance over the UV2 band. In the case of the wavelength dependent ISRF fit, the impact  
241 of first-order PAs on OMI radiances is relatively visible in the wavelength range of 300-310 nm. This result  
242 is physically consistent with the wavelength dependent property shown in the slit parameters derived from  
243 OMI irradiances as shown in Figure 6 where slit parameters are characterized in 10-pixel increments



244 assuming the super Gaussian slit function. In UV1, the slit widths plotted as FWHM slightly decrease by ~  
245 0.1 nm at shorter wavelengths than 288 nm, but more sharply vary by up to ~ 0.2 nm at longer wavelengths.  
246 Compared to slit widths, the wavelength dependences of the shape factors are less noticeable, except at  
247 boundaries of the window. In the UV2 window, both slit width and shape factor are highly invariant.

### 248 **3.2 Impact of including pseudo absorbers on ozone profile retrievals**

249 Figures 7 to 10 evaluate the impact of including zero-order PAs on ozone profile retrievals. Figure 7  
250 illustrates how different assumptions in the slit functions affect the ozone profile retrievals with respect to  
251 the retrieval sensitivity and the fitting accuracy from the case shown in Figure 4. In this figure, the Degrees  
252 of Freedom for Signal (DFS) represents the independent pieces of ozone information available from  
253 measurements, which typically decreases as ozone retrievals are further constrained by other fitting  
254 variables. The reduced DFS values (< 5 %) imply that the ozone retrievals are correlated slightly with PAs.  
255 The fitting accuracy is assessed as the root mean square (RMS) of relative difference (%) between measured  
256 and calculated radiances over the UV1 and UV2 ranges, respectively. Including the PAs makes little  
257 difference in the UV1 fitting residuals for most of individual pixels (1-5 %), but significantly reduces  
258 residuals in the UV2 range. The adjusted amount of the residuals with PAs are generally larger when  
259 assuming super Gaussian slit functions. This comes from different assumptions for slit functions in deriving  
260 soft calibration spectra, where slit functions were parameterized as standard Gaussians. Therefore, applying  
261 soft calibration to OMI spectra entails somewhat artificial spectral structures if ISRFs are assumed as Super  
262 Gaussian in ozone retrievals, and hence the impact of PAs on the spectral fitting becomes more considerable.  
263 Figure 8 compares how the spectral residuals are adjusted with PAs when soft calibration is turned on and  
264 off, respectively. Using super Gaussians causes larger amplitudes of the spectral fitting residuals than using  
265 standard Gaussians, if soft calibration is turned on and PAs are excluded. On the other hand, some residuals  
266 are reduced and more broadly structured if soft calibration is turned off. Including PAs eliminates/reduces  
267 some spikes of fitting residuals as well as improves the consistency of the fitting accuracy between using  
268 standard and super Gaussians at wavelengths above 300 nm.

269 The benefit of this implementation on ozone retrievals is further assessed through comparison with  
270 Electrochemical Concentration Cell (ECC) ozonesondes collected from the WOUDC (<https://woudc.org/>)  
271 and SHADOZ (<https://tropo.gsfc.nasa.gov/shadoz/>) networks during the period 2005 to 2008. We select 13  
272 SHADOZ sites in the tropics and 38 WOUDC sites in the northern mid/high latitudes. The collocation  
273 criteria is within +/- 1 ° in latitude and longitude and within 12 hours in time. For comparison, high-vertical  
274 resolution (~100 nm) profiles of ozonesondes are interpolated onto OMI retrieval grids (~2.5 km thick).  
275 We limit OMI/ozonesonde comparisons to OMI solar zenith angle < 85°, effective cloud fraction < 0.4,  
276 surface albedo < 20 % (100 %) in tropics and mid-latitudes (high latitude), top altitude of ozonesondes >



277 30 km, ozonesonde correction factors ranging from 0.85 to 1.15 if they exist, and data gaps for each  
278 ozonesonde no greater than 3km. Comparisons between OMI and ozonesondes are performed for the  
279 tropospheric ozone columns (TCOs) over 3 different latitude bands and for ozone profiles including all the  
280 sites, with and without PAs (zero-order) for standard and super Gaussian slit function changes, respectively.

281 Figure 9 shows the comparisons of tropospheric ozone columns as scatter plots. Without using PAs, the  
282 retrievals show significant differences of (1.2-2.2 DU or 3.8-6.4%) especially in mean biases between super  
283 and standard Gaussians, with negative biases of 0.2-0.7 DU for super Gaussians and positive biases of 0.8-  
284 1.5 DU for standard Gaussians. Overall, OMI retrievals are in a better agreement with ozonesonde  
285 measurements using super Gaussians. The correlations and standard deviations are very similar in the  
286 tropics and mid-latitudes, but the retrievals with standard Gaussians show better correlation and smaller  
287 standard deviations in high-latitudes. Consistent with Sun et al (2017), the retrievals show significant  
288 differences between using standard and super Gaussians, although there are some inconsistencies in  
289 comparing OMI and ozonesondes; the main inconsistent factors are listed as following: In this study, soft  
290 calibration is turned on and a priori information is taken from TB climatology to perform OMI ozone profile  
291 retrievals, whereas soft calibration is turned off and a priori information is taken from LLM climatology in  
292 Sun et al. (2017). OMI/ozonesonde data filtering criteria are quite similar to each other, except that the  
293 criteria of the solar zenith angle and cloud fraction are relaxed from 75° and 0.3 to 85° and 0.4, respectively,  
294 and the adjustment of ozonesondes with correction factor given for the WOUDC dataset is turned on in this  
295 study. Comparison is performed by latitudes here whereas global comparison is analyzed in Sun et al.  
296 (2017). After accounting for the slit differences between radiances and irradiances using PAs, the retrievals  
297 are significantly improved for both standard and super Gaussians and these two retrievals become consistent  
298 except for the use of super Gaussians in the tropics. The mean biases in the tropics and mid-latitudes are  
299 almost eliminated, to within 0.3 DU, but the standard deviations and correlation do not change much,  
300 slightly worse in the tropics and better in the mid-latitudes. In the high-latitudes, the standard deviations  
301 and correlation are significantly improved especially for using super Gaussians, but the mean biases are  
302 similar to the standard Gaussian without PAs. The lack of improvement with PAs in the tropics with super  
303 Gaussians illustrates that ISRFs of radiances are quite similar to those of irradiances in the tropics, while  
304 super Gaussians better parameterize OMI ISRFs than standard Gaussians. This is consistent with the  
305 comparison of the fitting accuracy of the UV2 band as shown in Figure 7, where the fitting residuals are  
306 slightly reduced in the tropics when super Gaussians are linearized, but the standard Gaussian linearization  
307 significantly improves the fitting accuracy. The mean biases of the profile comparison as shown in Figure  
308 10 clearly shows that including PAs to account for ISRF differences significantly reduces mean biases  
309 below 10 km and the general altitude dependence and improves the consistency between using standard  
310 and super Gaussians; the standard deviations also show noticeable improvement in the altitude range of 10-



311 20 km for both Gaussians. The significant improvement at all latitudes corroborates the change of ISRFs  
312 between radiance and irradiance along the orbit as conjectured by Sun et al. (2017). The consistency  
313 between using standard and super Gaussians after using PAs is mainly because there is strong anti-  
314 correlation between the slit width and shape partial derivatives as shown in Figure 2, so the adjustment of  
315 slit width only in the use of standard Gaussian can achieve almost the same effect as the adjustment of both  
316 parameters in the use of super Gaussian. Accounting for the wavelength dependent change of the ISRFs  
317 with first-order PAs makes insignificant differences to both fit residuals and ozone retrievals (not shown  
318 here). This could be mainly explained with the fact of the negligible wavelength dependence of OMI ISRFs  
319 especially in UV2 as shown in Figure 5 where the PA spectrum ( $\frac{\partial \ln I}{\partial p} \cdot \Delta p$ ) shows almost no variance,  
320 except at the upper boundary of the UV1 as well as in Figure 6 where the UV2 slit parameters derived from  
321 irradiances in the sub-fit windows vary within 0.05 nm for FWHM and 0.2 for shape factor.

322

#### 323 **4. Summary**

324 The knowledge of the Instrument Spectral Response Functions (ISRFs) or slit functions is important  
325 for ozone profile retrievals from the Hartley and Huggins bands. ISRFs can be measured in the laboratory  
326 prior to launch, but they have been typically derived from solar irradiance measurements assuming  
327 Gaussian-like functions in order to account for the effect of the ISRF changes after launch. However, the  
328 parameterization of the ISRFs from solar irradiances could be inadequate for achieving a high accuracy of  
329 the fitting residuals as ISRFs in radiances could significantly deviate from those in solar radiances (Beirle  
330 et al., 2017) and might affect ozone profile retrievals as suggested in Sun et al. (2017). Therefore, this study  
331 implements a linearization scheme to account for the spectral errors caused by the ISRFs changes as Pseudo  
332 Absorbers (PAs) in an optimal estimation based fitting procedure for retrieving ozone profiles from OMI  
333 BUV measurements using the SAO ozone profile algorithm. The ISRFs are assumed to be the generic super  
334 Gaussian that can be used as standard Gaussian when fixing the shape factor to 2. This linearization was  
335 originally introduced in Beirle et al. (2017) for DOAS analysis, but this study extends this application and  
336 more detail how to implement in practice using two different approaches to derive radiance errors from slit  
337 function partial derivatives with respect to slit parameters. These two approaches correspond to the two  
338 methods of simulating radiances at instrument spectral resolution, one using effective cross sections which  
339 were previously used in the SAO ozone profile algorithm and are still used in most of the trace gas retrievals  
340 from the UV and visible, and the other calculating radiances at high resolution before convolution, which  
341 is the preferred method in the SAO ozone profile algorithm. Consistent PAs are derived with these two  
342 approaches, as expected.



343 The fitting coefficients ( $\Delta p$ ) to the PAs, representing the difference of slit parameters between radiance  
344 and irradiance, are iteratively fitted as part of the state vector along with ozone and other parameters. The  
345 UV1 slit parameters show distinct cross-track-dependent differences, especially in high-latitudes. In  
346 addition, an abnormal  $\Delta p$  caused by scene heterogeneity is observed around bright surfaces and cloudy  
347 scenes. The total PA spectrum ( $\frac{\partial I}{\partial p} \cdot \Delta p$ ) illustrates that the slit width change causes most of the spectral  
348 structures in the UV1 band because the OMI ISRFs are close to Gaussian. Otherwise, the ISRF change  
349 results into different spectral responses in the UV2 band with different Gaussian functions because the  
350 adjustment of the shape factor becomes more important in accounting for the convolution error when using  
351 super Gaussians.

352 Insignificant wavelength dependence on OMI slit functions is demonstrated from slit function  
353 parameters derived from irradiances in the sub-fit window, which leads to little difference in ozone profile  
354 retrievals when zero and first-order wavelength dependent PA coefficients are implemented to fit the  
355 spectral structures caused by slit function errors, respectively. Therefore we evaluate the benefit of  
356 including the zero-order PAs fit on both the accuracy of the fitting residuals and the quality of retrieved  
357 ozone profiles through validation against ozonesonde observations. Some spikes in the fitting residuals are  
358 reduced or eliminated. Commonly, including PAs makes little change on both fit residuals and ozone  
359 retrievals in the tropics if a super Gaussians are assumed as ISRFs but this is not the case for the standard  
360 Gaussian assumption. Retrievals using standard and super Gaussians agree better if slit function errors are  
361 accounted for by including PAs. Using PAs ultimately demonstrates substantial improvement of ozone  
362 profile retrievals in the comparison of tropospheric ozone columns and ozone profiles up to 30 km. Using  
363 super Gaussians, the TCO comparison shows significant improvement in mean biases in mid-latitudes and  
364 in standard deviations in high-latitudes. Using standard Gaussians, the TCO comparison also shows  
365 significant improvement in mean biases in the tropics. The profile comparison generally shows  
366 improvement in mean biases as well as in standard deviation in the altitude range 10-20 km. More  
367 importantly, using these PAs make the retrieval consistent between standard and super Gaussians. Such  
368 consistency is due to the anti-correlation between slit width and shape PAs. This study demonstrates the  
369 slit function differences between radiance and irradiance and its usefulness to account for such differences  
370 on the pixel-to-pixel basis. In this experiment, the soft spectrum, derived with the standard Gaussian  
371 assumption, is applied to remove systematic measurement errors before spectral fitting, indicating that the  
372 evaluation of ozone retrievals might be unfairly performed for the super Gaussian function implementation.  
373 Nonetheless, OMI ozone profile retrievals show better agreement with ozonesonde observations when the  
374 super Gaussian is linearized. Actually, the fitting residuals are slightly more broadly structured with super  
375 Gaussians than with standard Gaussians if the soft-calibration and PAs are turned off, indicating the benefit



376 of deriving a soft calibration with the super Gaussians. Therefore, there is still room for achieving better  
377 benefits when using the PAs on ozone profile retrievals by applying the soft calibration derived with super  
378 Gaussians.

379

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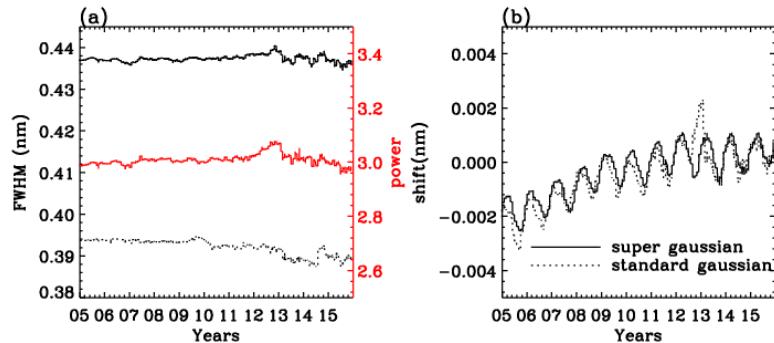
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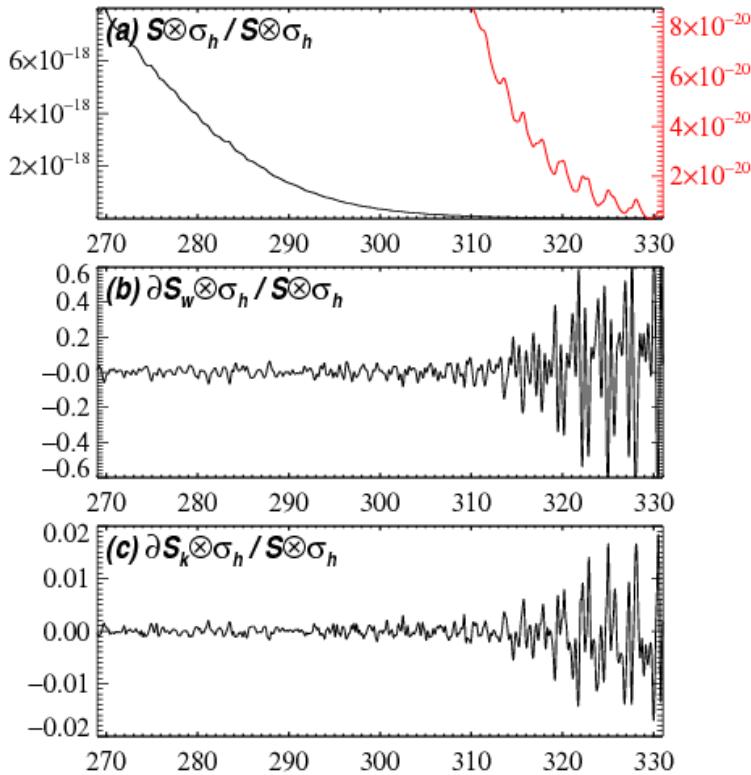
450 **Figure 1.** Time series of (a) slit parameters and (b) wavelength shifts for OMI daily irradiance  
451 measurements (310-330 nm) at nadir cross track position when Super Gaussians (solid line) and  
452 standard Gaussians (dotted line) are parameterized as slit function shapes, respectively.

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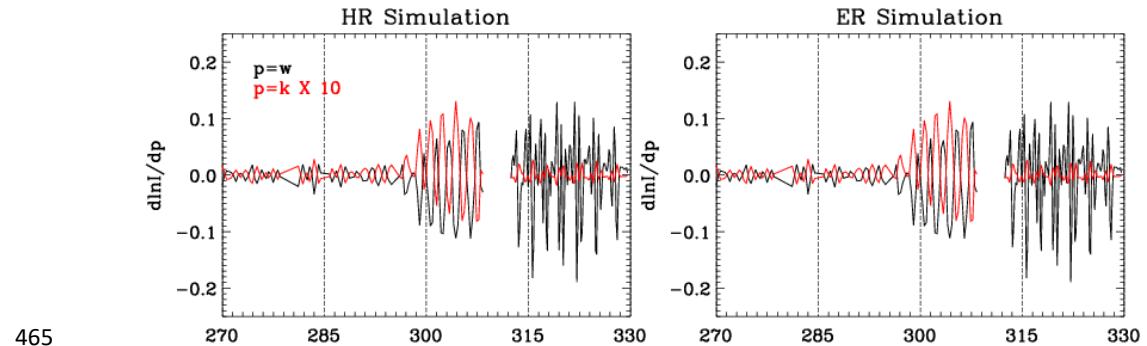


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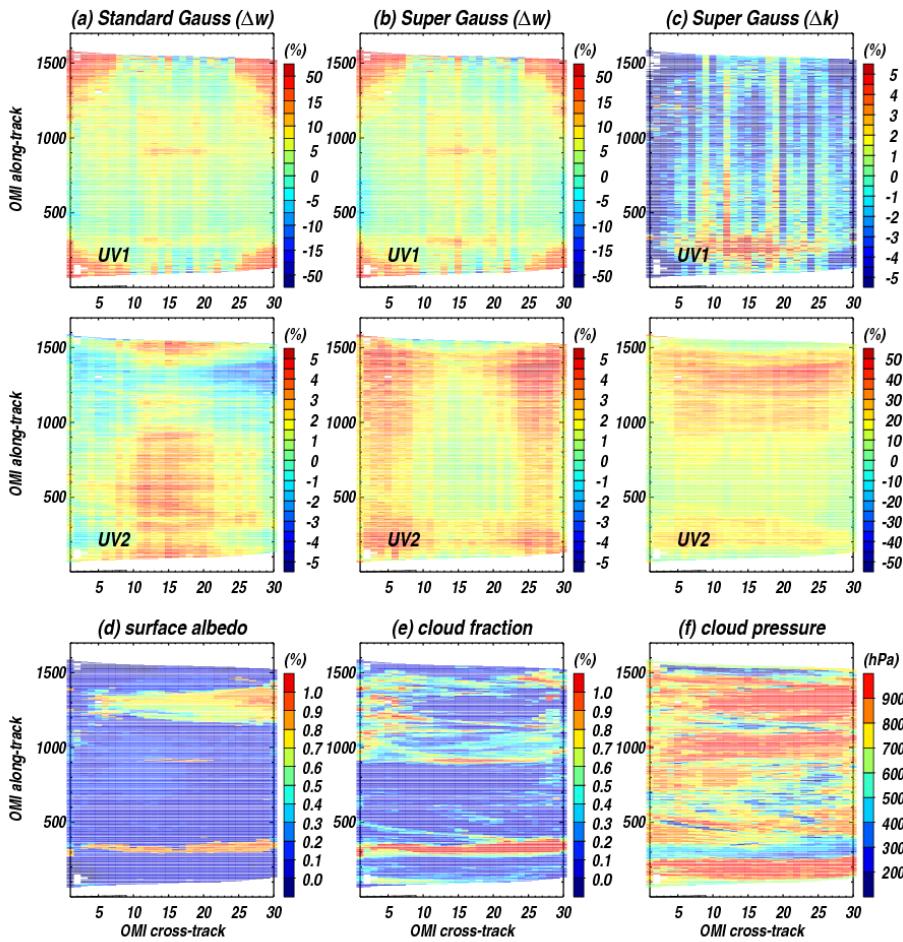
458 **Figure 2. (a)** Ozone absorption cross sections ( $\text{cm}^2/\text{molecule}$ ) ( $\delta_h$ ) at different scales (red and black) at  
459 a representative temperature (238.12 K) calculated via convolution of high-resolution (0.01 nm)  
460 reference spectrum with the Super Gaussian slit function,  $S$  ( $k = 2.6, w = 0.26 \text{ nm}$ ). **(b)** and **(c)** its  
461 derivatives with respect to slit parameters ( $\partial S_p = \frac{\partial S}{\partial p}$ ),  $w$  and  $k$ , respectively, normalized to the  
462 convolved cross sections.

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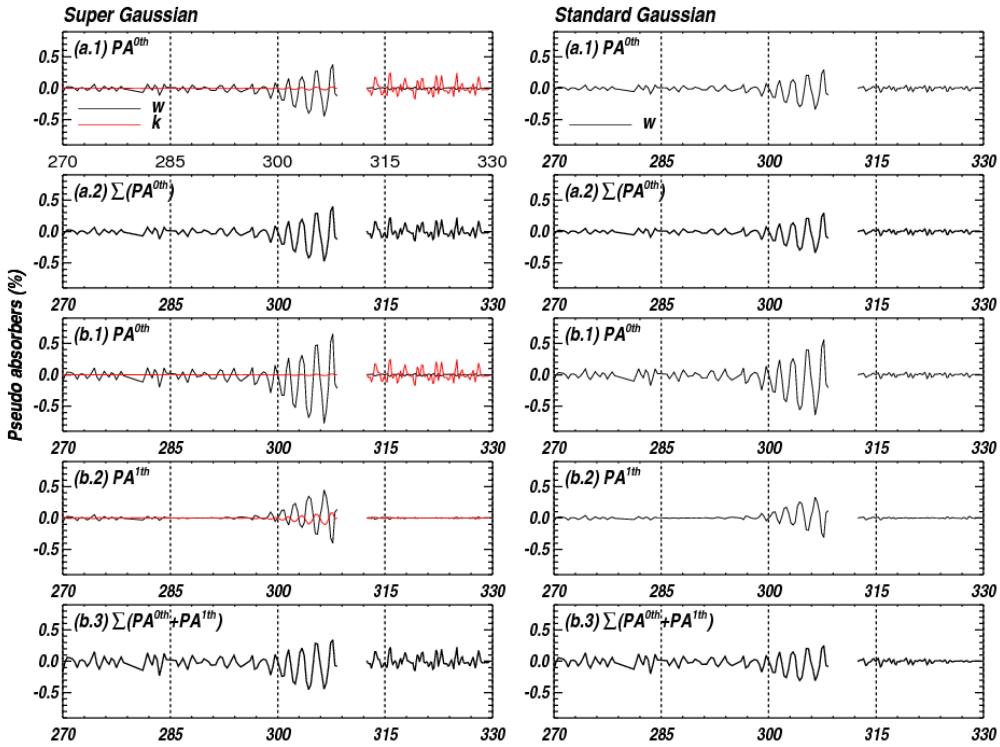


465  
466 **Figure 3. Derivatives of OMI radiance spectrum simulated using high-resolution (HR) and effective**  
467 **resolution (ER) cross section spectra with respect to slit parameters assuming a Super Gaussian function.**  
468 **dlnI/dk is multiplied by a factor of 10 to visually match dlnI/dw in the same y-axis.**



469

470 **Figure 4. Pseudo absorption coefficients ( $\Delta w, \Delta k$ ) for fitting the OMI radiances due to slit function**  
 471 **changes assuming (a) standard Gaussian and (b-c) Super Gaussian, within the first orbit of**  
 472 **measurements on 1 July 2006, with (d-f) the corresponding geophysical parameters.  $\Delta w$  and  $\Delta k$  is**  
 473 **displayed after being normalized with  $w_o$ , and  $k_o$ , the slit parameters derived from OMI solar**  
 474 **irradiance measurements.**



475  
 476 **Figure 5. (a.1)** Pseudo absorber spectra ( $\frac{\partial \ln I}{\partial p} \times \Delta p$ ) for zero order slit parameters and **(a.2)** its total  
 477 spectra for (left) Super Gaussian and (right) Standard Gaussian function parameterizations, respectively.  
 478 **(b)** Same as (a), but for first order polynomial fit. The case represents an average at nadir in the latitude  
 479 zone 30°-60°N.

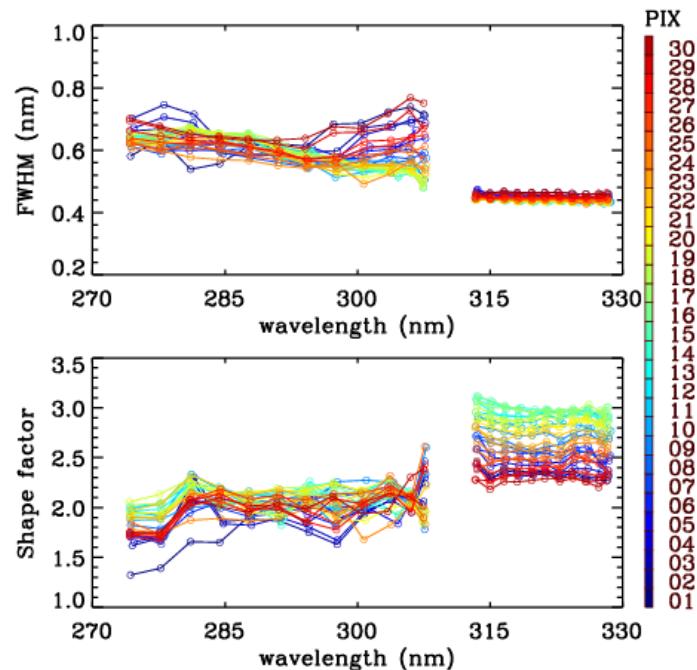
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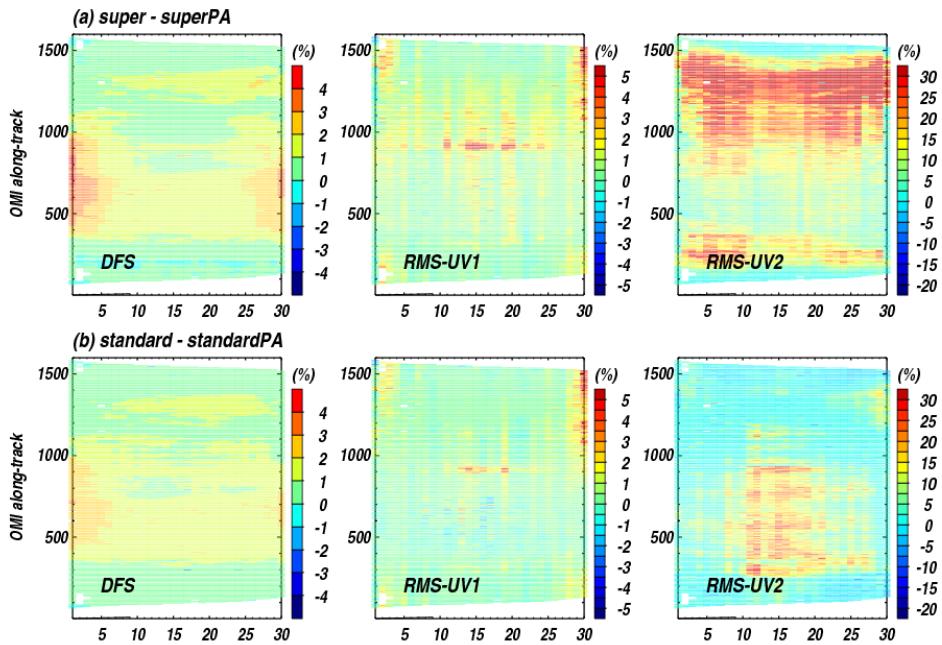
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486 **Figure 6. OMI ISRF FWHM (nm) and shape factor ( $k$ ) as functions of the center wavelength, as derived**  
487 **from OMI solar irradiances assuming Super Gaussian functions over a range of 31 spectral pixels in 10-**  
488 **pixel increments. Different colors represent different cross-track positions from 1 (blue) to 30 (red).**



489

490      **Figure 7.** Same as Figure 4, but for comparisons of the Degrees of Freedom for Signal (DFS) and the Root  
491      Mean Square (RMS) of spectra fitting residuals in UV 1 and UV2 with and without zero-order pseudo  
492      absorber. Positive values indicate that both fitting residuals and DFSs are reduced due to the pseudo  
493      absorber.

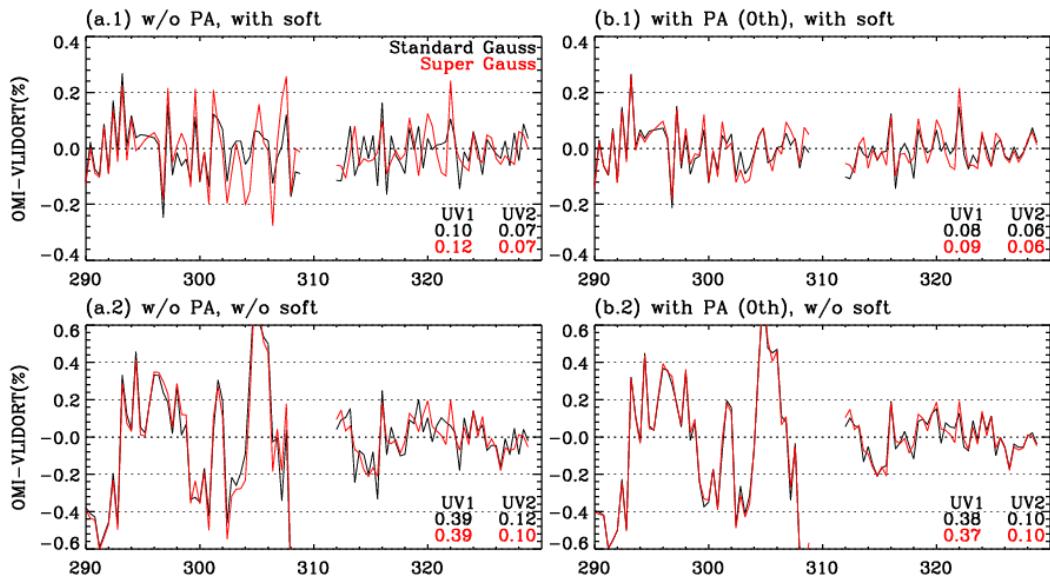
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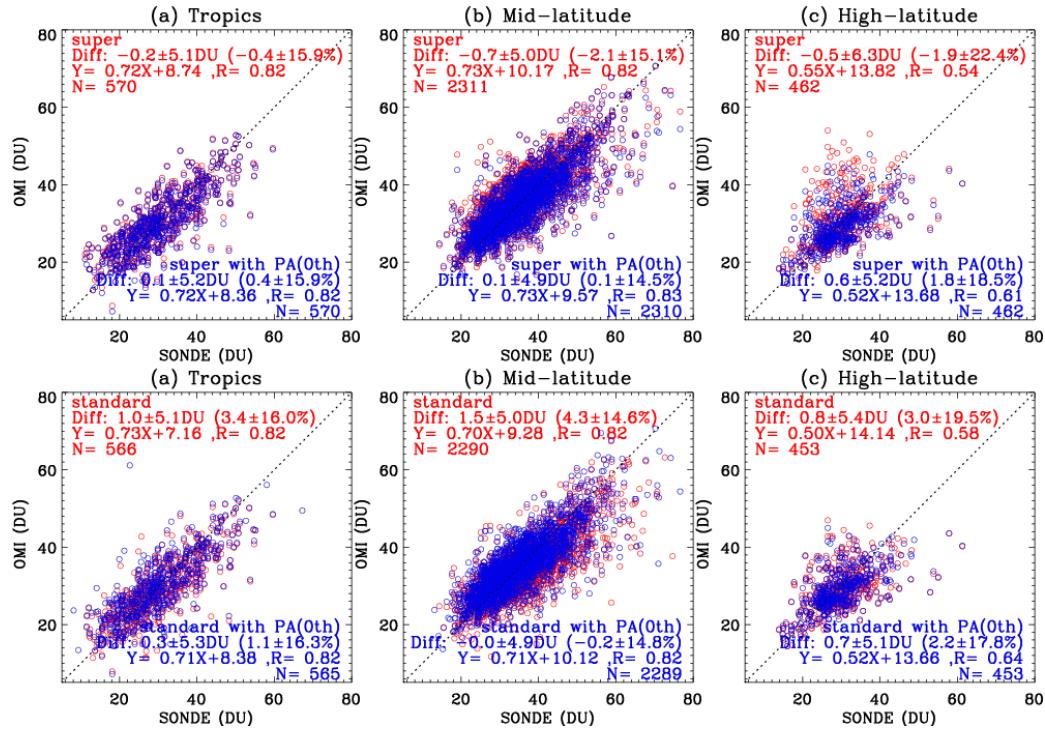
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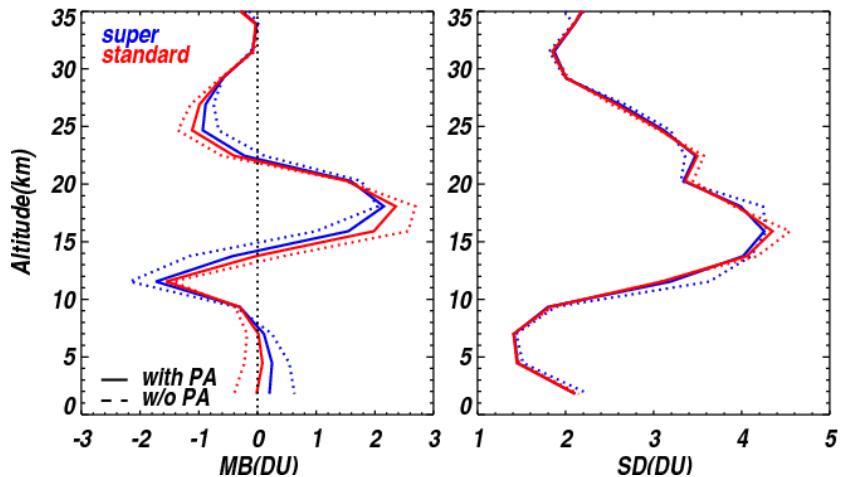
500 **Figure 8. Average differences (%) between measured (OMI) and simulated (VLIDORT) radiances**  
501 **(residuals) at the nadir cross-track pixel in the tropics (30°S-30°S) without (a) and with (b) zero-order**  
502 **pseudo absorbers (PA) when the standard Gaussian (black line) and the Super Gaussian (red line) are**  
503 **assumed as ISRFs, respectively. Upper/lower panels represent the fit results with soft calibration being**  
504 **turned on/off. The residuals in the UV1 (< 310 nm) are scaled by a factor of 2 to fit in the given y-axis. In**  
505 **the legend, the RMS of residuals (%) are given for UV1 and UV2 wavelength ranges, respectively.**

506

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517 **Figure 10. Global mean biases at each OMI layer and  $1\sigma$  standard deviations of the differences between**  
518 **OMI and ozonesondes, with different slit function assumptions/implementations.**

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522