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Supplement of

A new discrete wavelength backscattered ultraviolet algorithm for consistent volcanic SO_2 retrievals from multiple satellite missions

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Supplement of Discrete wavelength SO_2 Algorithms Fisher et al.

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S1. TOMS instrument description

The TOMS is a fixed-grating Ebert-Fastie monochromator with a photomultiplier tube detector that measures solar backscattered ultraviolet (BUV) radiances (I_m) at six narrow wavelengths bands (Full Width at Half Maximum band width ~1.1nm) in the near ultraviolet (UV) spectral region, as well as the incident solar irradiances (F) (Heath, *et al*., 1975). The ratio of radiance to irradiance provides the spectral reflectivity parameter used in the retrieval. The wavelength band centers shown in Table 2 were selected to optimize column ozone retrievals assuming that two pairs of shorter, absorbing wavelengths would be needed to cover the full dynamic range of ozone and solar zenith angles encountered globally, similar to the Dobson Spectrophotometer design. Two additional non-absorbed longer wavelengths were provided to measure the surface or cloud reflectivity (R) and its spectral dependence. TOMS scans in the cross-track direction in 3° steps from 51° on west side of nadir to 51° on the east, for a total of 35 cross-track samples. The instantaneous field-of-view (IFOV) of 3° x 3° results in a effective field-of-view (EFOV) varying from a 50 km x 50 km square FOV at nadir to a 125 km x 280 km diamond-shaped FOV at the scan extremes. The total swath width is 3000 km covering Earth's surface in 14 orbits per day.

S3. New MS_SO2 algorithm

The MS_SO2 forward model uses LUTs produced using the Radiative Transfer Model (TOMRAD). For a given wavelength (λ), viewing geometry (θ_0, θ_S, ϕ), atmospheric pressure (P), standard ozone profile (Ω_P), SO_2 Gaussian profile (Σ_P), surface reflectivity (R_S) a satellite-observed BUV radiance, I_c, is determined from five computed BUV radiance parameters, I_0 (black surface BUV), atmospheric directional transmittivity (T) and the spherical reflectivity of the atmosphere (S_b), assuming Lambertian reflecting surface :

$$I_c(\lambda, \theta_0, \theta_S, \phi, P, \Omega_P, \Sigma_P, R_S) = I_0 + I_1\cos(\phi) + I_2\cos(2\phi) + \frac{R_S T}{(1-R_S s_b)}, \quad \text{(S1)}$$

The LUTs include two additional terms Z1 and Z2, from which I_1 and I_2 are defined:

$$Z_1 = \frac{I_1}{-(3/8)\mu_0\sqrt{(1-\mu_0^2)(1-\mu^2)}} \quad \text{(S2a)}$$

$$Z_2 = \frac{I_2}{-(3/32)\mu_0\sqrt{(1-\mu_0^2)(1-\mu^2)/\mu}} \quad \text{(S2b)}$$

where μ_0=cosθ_0 and μ=cosθ_s and θ_0 and θ_s are the solar and satellite viewing zenith angles. Z1 and Z2 improve interpolations between LUT nodes. The calculated BUV radiance I_c is subsequently converted to an N-value, as defined in Eq. (1).

S3.1 Step 1 retrieval

Equation S1 shows the 4x4 K-matrix consists of weighting factors corresponding to the Jacobians of the 4 retrieved parameters**.** The columns of K correspond to the retrieved parameters described in Eq. 4, while the rows correspond to the four used radiances from the TOMS instrument (317, 331, 340 and 380 nm)

$$\mathbf{K} = \begin{pmatrix} \frac{\partial N_{317}}{\partial \Omega} & \frac{\partial N_{317}}{\partial \Sigma} & \frac{\partial N_{317}}{\partial R}(\lambda_1 - \lambda_R) & \frac{\partial N_{317}}{\partial R} \\ \frac{\partial N_{331}}{\partial \Omega} & \frac{\partial N_{331}}{\partial \Sigma} & \frac{\partial N_{331}}{\partial R}(\lambda_2 - \lambda_R) & \frac{\partial N_{331}}{\partial R} \\ \frac{\partial N_{340}}{\partial \Omega} & \frac{\partial N_{340}}{\partial \Sigma} & \frac{\partial N_{340}}{\partial R}(\lambda_3 - \lambda_R) & \frac{\partial N_{340}}{\partial R} \\ 0 & 0 & 0 & \frac{\partial N_{380}}{\partial R} \end{pmatrix} \quad \text{(S3)}$$

The reflectivity R_S is computed using the measurement I_m and parameters I_0, T and s_b from the radiance LUTs defined described in the previous section:

$$R_s = \frac{(I_m - I_0)}{T + s_b((I_m - I_0))}. \quad \text{(S4)}$$

S3.2 Step 2 Procedure to correct ozone inside SO_2 plume

The ozone sample does not include Ω for any scenes inside the plume or that exceed the highest nodal point in the ozone table for a given latitude band. The corrected ozone is subsequently treated as a constant, which fixes the total ozone with respect to the LUT total ozone nodes (the O_3 is free to vary up and down in Step 1 while iteratively converging on a solution). This sample of unperturbed ozone values is next further divided into two sub-samples spanning 30° north and south of the target FoV.

Linear regressions are then performed on each subsample, using the latitude coordinate along the orbit as the independent variable. This method elicits two sets of regression parameters north and south of the target FoV. These parameters are used in Eqs. (5a,b) to obtain two values, Ω_1 and Ω_2, at the position of the target pixel, $\varphi_{\Omega 0}$:

$$\Omega_1 = m_1 \varphi_{\Omega 0} + b_1 \quad \text{(S5a)}$$

$$\Omega_2 = m_2 \varphi_{\Omega 0} + b_2 \quad \text{(S5b)}$$

To determine the corrected ozone, Ω_1 and Ω_2 are then weighted by the relative distance to the pixel in each subsample closest to the plume boundary, d_1 and d_2, as shown in Eq (S6,a,b):

$$w_1 = \frac{d1}{d1 + d2} \quad \text{(S6a)}$$

$$w_2 = \frac{d2}{d1 + d2}. \quad \text{(S6b)}$$

These weights together with Ω_1 and Ω_2 determine the corrected ozone value, Ω_{cor}, as \shown,

$$\Omega_{cor} = w_1\Omega_1 + w_2\Omega_2. \tag{S7}$$

S3.3 Soft Calibration: N-value bias correction

The entire TOMS data record was processed with MS_SO2 and a small retrieval bias was detected in the global SO_2 background. We subsequently developed a procedure to correct for this bias based on a technique described by Rogers (2000). For this procedure, we first construct a matrix G defined as the inverse of K defined as,

$$G = K^{-1}. \tag{S8}$$

The G matrix characterizes the effects on the retrieved variable, x_i, due to small perturbations of the measured N-value. The matrix elements of G are shown in Fig. S2 for O_3 and SO_2 at the three absorbing wavelengths at 317, 331 and 340 nm as a function of latitude. The elements for O_3 and SO_2 are anti-correlated at each wavelength, exhibit a strong dependence on the viewing geometry and peak at the equator (i.e., absolute value peaks). The change in gas amount is inversely related to the gas sensitivity. For example, a 1 N-value change at 331 nm results in a 100 DU change in O_3, whereas the same 1 N-value change at 317 nm only results in about a -1 DU change, a two order of magnitude difference. Consequently, a small perturbation at 340 nm has a much larger effect on the retrieved SO2 than at 317 nm.

According to Rogers (2000), the error in the retrieval can defined by the action of G on the measurement error, ε_y, as described by Eq. (S9),

$$dx = G\varepsilon_y. \tag{S9}$$

The retrieval bias estimated from an analysis of the global background is corrected for by shifting the whole distribution shown in Fig. 9b so that the mean of the distribution is zero. We adapted soft calibration procedure that computes a small correction to a single channel. The 340 nm channel was selected because it is the least sensitive absorbing channel with respect to O_3 and SO_2 absorption.

Eq. (S9) can then be expressed as,

$$d\Sigma = \frac{\partial\Sigma}{\partial N_{340}} \mathrm{dN}_{340}. \tag{S10}$$

where $\partial\Sigma/\partial N_{340}$ and $d\Sigma$ represents the G matrix element for SO_2 at 340 nm and the SO_2 retrieved in the SO_2 free regions of the atmosphere, respectively.

The correction to the measured radiance dN_{340} can be interpreted as the change in N_{340} needed to shift the distribution in Fig. 8b so that $\langle\Sigma\rangle = 0$. For this analysis, the same 90 orbits described in Section 5.1 were selected. The Step 1 Σ retrieved between 60 S and 60ºN was used to calculate dN_{340} as a function of cross track position. Equation (19) is then solved for dN_{340} with respect to a large sample as a function of the swath position,

$$dN_{340} = Mean\left(\frac{d\Sigma}{\frac{\partial \Sigma}{\partial N_{340}}}\right) \quad \text{(S11)}$$

This correction factor was first determined off-line and then applied to the entire data record during operational processing. Figure S3 shows the dN_{340} correction as a function of the TOMS cross-track (swath) position. Overall the correction is small ~0.1 and practically does not depend on the swath position.

S4.2.1 Uncertainties due to SO_2 plume height

S5.1 Mount Pinatubo

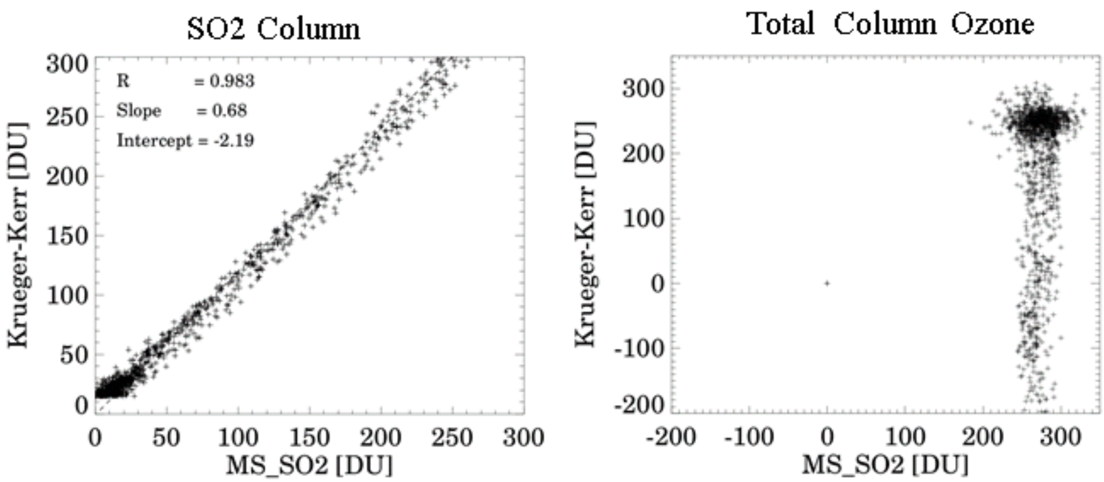

Table S1. Percentage error in SO_2 Jacobians at 317 nm due to a 2-km error in the assumed plume height for typical observation conditions in the tropics (30°S-30°N, SZA=10°, RAZ=90°, O_3 = 275 DU).

SO_2 (DU)	13 km *a priori* profile*				18-km *a priori* profile**			
	VZA=0°		VZA=60°		VZA=0°		VZA=60°	
	R=0.05	R=0.50	R=0.05	R=0.50	R=0.05	R=0.50	R=0.05	R=0.50
50	5.4 (-3.0)	0.6 (0.1)	9.7 (-5.9)	4.5 (-2.7)	1.3 (-0.8)	-0.5 (0.4)	3.4 (-2.3)	1.4 (-0.9)
100	8.7 (-5.4)	3.1 (-1.9)	14.2 (-9.1)	8.2 (-5.5)	3.1 (-2.2)	1.0 (-0.7)	6.0 (-4.2)	3.5 (-2.5)
200	14.6 (-9.4)	7.5 (-5.1)	22.1 (-14.4)	15.2 (-10.3)	6.2 (-4.3)	3.2 (-2.2)	11.0 (-7.7)	7.4 (-5.3)
300	20.1 (-13.0)	12.0 (-8.1)	28.3 (-18.6)	21.8 (-14.8)	9.3 (-6.4)	5.4 (-3.8)	16.2 (-11.4)	12.0 (-8.6)

*Error is calculated as the relative percentage difference in SO_2 Jacobians between the profile with a center mass altitude (CMA) of 11 km (15 km for results in parentheses) and the assumed *a*

priori profile with a CMA of 13 km. A positive (negative) error means SO_2 Jacobians are overestimated (underestimated) with the *a priori* profile and the retrieved SO_2 column amount will be biased low (high).
**Error is calculated as the relative percentage difference in SO_2 Jacobians between the profile with a CMA of 16 km (20 km for results in parentheses) and the assumed *a priori* profile with a CMA of 18 km.

Table S2. Percentage error in SO_2 Jacobians at 317 nm due to a 2-km error in assumed plume height for typical observation conditions in the mid latitudes (30-60°N/S, SZA=40°, RAZ=90°, O_3 = 325 DU).

SO_2 (DU)	13 km *a priori* profile*				18-km *a priori* profile**			
	VZA=0°		VZA=60°		VZA=0°		VZA=60°	
	R=0.05	R=0.50	R=0.05	R=0.50	R=0.05	R=0.50	R=0.05	R=0.50
50	6.5 (-4.1)	1.5 (-0.9)	10.8 (-7.0)	6.0 (-4.1)	2.4 (-1.8)	0.5 (-0.4)	4.6 (-3.5)	2.7 (-2.1)
100	10.2 (-6.7)	4.4 (-3.0)	15.8 (-10.5)	10.3 (-7.2)	4.4 (-3.2)	2.0 (-1.5)	7.6 (-5.5)	5.1 (-3.8)
200	16.8 (-11.0)	9.6 (-6.6)	24.1 (-15.9)	18.1 (-12.5)	7.9 (-5.7)	4.6 (-3.4)	13.3 (-9.6)	9.9 (-7.3)
300	22.7 (-14.9)	14.9 (-10.2)	29.4 (-19.8)	24.4 (-16.9)	11.8 (-8.4)	7.5 (-5.4)	18.7 (-13.6)	15.2 (-11.2)

*See footnote for Table 1.
**See footnote for Table 1.

Table S3. Percentage error in SO_2 Jacobians at 317 nm due to a 2-km error in assumed plume height for typical observation conditions in the high latitudes (60-90°N/S, SZA=60°, RAZ=90°, O_3 = 375 DU).

SO_2 (DU)	13 km *a priori* profile*				18-km *a priori* profile**			
	VZA=0°		VZA=60°		VZA=0°		VZA=60°	
	R=0.05	R=0.50	R=0.05	R=0.50	R=0.05	R=0.50	R=0.05	R=0.50
50	10.1 (-7.0)	4.6 (-3.5)	14.5 (-10.0)	10.0 (-7.4)	5.2 (-4.0)	2.8 (-2.3)	7.9 (-6.2)	5.9 (-4.7)
100	14.5 (-10.0)	8.4 (-6.2)	20.0 (-13.7)	15.2 (-10.9)	7.6 (-5.8)	4.7 (-3.7)	11.6 (-8.8)	9.1 (-7.1)
200	21.8 (-14.8)	15.0 (-10.7)	27.4 (-18.8)	23.0 (-16.4)	12.6 (-9.4)	8.7 (-6.7)	18.3 (-13.8)	15.5 (-11.9)
300	26.9 (-18.4)	20.7 (-14.8)	29.9 (-21.0)	26.8 (-19.4)	17.6 (-13.1)	13.3 (-10.1)	23.0 (-17.6)	20.7 (-16.0)

*See footnote for Table 1.
**See footnote for Table 1.

Table S4. Percentage errors in SO_2 (εSO2) and O_3 (εO3) total column amounts and *N* value residuals at 312 nm (Res_{312}) due to the use of *a priori* profile that is 2 km too high (center mass altitude: *a priori*: 13 km, actual: 11 km).

Tropics (O3=275 DU, SZA=10), Actual Profile: 11 km, *a priori*: 13 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO_2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	-4.0	0.1	-0.2	0.8	0.1	-0.2	-7.7	0.1	-0.4	-2.8	0.2	-0.4
100	-5.9	0.3	-0.7	-0.7	0.4	-0.7	-10.5	0.5	-1.2	-5.1	0.5	-1.3
200	-9.6	1.2	-2.0	-3.5	1.1	-1.9	-16.4	1.7	-3.2	-9.6	1.5	-3.4

300	-13.4	2.5	-3.6	-6.2	2.2	-3.6	-23.0	3.8	-5.1	-14.9	3.3	-5.5
Mid latitudes (O3=325 DU, SZA=40), Actual Profile: 11 km, *a priori*: 13 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO_2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	-4.7	0.1	-0.3	0.1	0.1	-0.3	-8.4	0.1	-0.5	-4.0	0.1	-0.5
100	-7.0	0.3	-0.8	-1.7	0.3	-0.8	-11.7	0.5	-1.3	-6.7	0.4	-1.4
200	-11.3	1.1	-2.2	-5.0	1.0	-2.3	-18.6	1.7	-3.0	-12.3	1.5	-3.2
300	-15.8	2.4	-3.8	-8.3	2.1	-3.9	-26.2	3.7	-4.4	-18.7	3.3	-4.7
High latitudes (O3=375 DU, SZA=60), Actual Profile: 11 km, *a priori*: 13 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO_2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	-7.8	0.1	-0.4	-2.8	0.1	-0.5	-11.6	0.2	-0.6	-7.5	0.2	-0.6
100	-10.7	0.4	-1.1	-5.1	0.3	-1.1	-15.9	0.5	-1.3	-11.2	0.5	-1.4
200	-16.6	1.3	-2.5	-9.7	1.1	-2.7	-25.0	1.9	-2.6	-19.3	1.7	-2.7
300	-23.2	2.8	-3.7	-14.9	2.4	-4.0	-35.1	4.2	-3.5	-28.6	3.9	-3.7

Table S5. Percentage errors in SO_2 (εSO2) and O_3 (εO3) total column amounts and *N* value residuals at 312 nm (Res_{312}) due to the use of *a priori* profile that is 2 km too low (center mass altitude: *a priori*: 13 km, actual: 15 km).

Tropics (O3=275 DU, SZA=10), Actual Profile: 15 km, *a priori*: 13 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	1.6	-0.1	0.2	-1.5	-0.1	0.2	4.8	-0.1	0.4	1.2	-0.1	0.4
100	3.5	-0.2	0.6	-0.1	-0.2	0.5	7.7	-0.4	1.2	3.3	-0.3	1.1
200	7.0	-1.0	1.9	2.2	-0.7	1.6	14.9	-1.7	3.6	7.8	-1.2	3.4
300	11.3	-2.3	3.9	4.6	-1.6	3.3	28.9	-5.1	6.6	14.9	-3.2	6.0
Middle latitudes (O3=325 DU, SZA=40), Actual Profile: 15 km, *a priori*: 13 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	2.8	-0.1	0.3	-0.5	-0.1	0.3	6.0	-0.1	0.5	2.6	-0.1	0.5
100	4.9	-0.3	0.8	1.0	-0.2	0.8	9.4	-0.4	1.4	5.1	-0.3	1.4
200	9.2	-1.0	2.4	3.7	-0.7	2.1	18.9	-1.8	3.7	11.2	-1.3	3.5
300	15.0	-2.4	4.4	6.9	-1.6	4.0	41.2	-6.3	6.1	22.5	-3.8	5.7
High latitudes (O3=375 DU, SZA=60), Actual Profile: 15 km, *a priori*: 13 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		

SO2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	6.3	-0.1	0.5	2.3	-0.1	0.5	10.1	-0.2	0.8	6.6	-0.1	0.7
100	9.2	-0.3	1.3	4.4	-0.3	1.2	15.4	-0.6	1.7	10.5	-0.4	1.7
200	16.8	-1.4	3.1	9.0	-1.0	3.0	35.2	-2.9	3.7	23.4	-2.1	3.6
300	31.4	-4.1	5.0	16.3	-2.5	4.8	N/A*	-20.8	5.5	N/A*	-16.8	7.6

*Solution did not converge after 20 iterations.

Table S6. Percentage errors in SO_2 (εSO2) and O_3 (εO3) total column amounts and *N* value residuals at 312 nm (Res_{312}) due to the use of *a priori* profile that is 2 km too high (center mass altitude: *a priori*: 18 km, actual: 16 km).

Tropics (O3=275 DU, SZA=10), Actual Profile: 16 km, *a priori*: 18 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	0.1	0.0	-0.1	1.6	0.0	-0.2	-1.8	0.1	-0.3	0.0	0.1	-0.3
100	-1.2	0.2	-0.4	0.6	0.2	-0.4	-3.5	0.2	-0.8	-1.4	0.2	-0.8
200	-3.2	0.6	-1.3	-0.9	0.5	-1.1	-6.7	0.9	-2.5	-3.8	0.7	-2.4
300	-5.1	1.3	-2.6	-2.3	1.0	-2.3	-10.3	2.0	-4.3	-6.5	1.6	-4.3

Middle latitudes (O3=325 DU, SZA=40), Actual Profile: 16 km, *a priori*: 18 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	-1.3	0.1	-0.3	0.5	0.1	-0.3	-3.1	0.1	-0.5	-1.4	0.1	-0.5
100	-2.6	0.2	-0.7	-0.5	0.2	-0.6	-5.0	0.3	-1.2	-2.9	0.2	-1.2
200	-4.7	0.6	-1.8	-2.1	0.5	-1.7	-8.7	1.0	-2.9	-5.8	0.8	-2.9
300	-7.1	1.3	-3.4	-3.7	1.0	-3.2	-13.3	2.2	-4.4	-9.4	1.7	-4.5

High latitudes (O3=375 DU, SZA=60), Actual Profile: 16 km, *a priori*: 18 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	-4.1	0.1	-0.5	-1.9	0.1	-0.5	-6.3	0.1	-0.8	-4.5	0.1	-0.8
100	-5.6	0.2	-1.2	-3.0	0.2	-1.1	-8.7	0.4	-1.7	-6.5	0.3	-1.7
200	-8.8	0.8	-2.8	-5.4	0.6	-2.8	-14.3	1.3	-3.3	-11.3	1.1	-3.4
300	-12.7	1.8	-4.3	-8.3	1.3	-4.3	-21.6	3.0	-4.2	-17.6	2.5	-4.4

Table S7. Percentage errors in SO_2 (εSO2) and O_3 (εO3) total column amounts and N value residuals at 312 nm (Res_{312}) due to the use of *a priori* profile that is 2 km too low (center mass altitude: *a priori*: 18 km, actual: 20 km).

Tropics (O3=275 DU, SZA=10), Actual Profile: 20 km, *a priori*: 18 km				
	VZA=0		VZA=60	
	R=0.05	R=0.50	R=0.05	R=0.50

SO2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	-0.3	0.0	0.1	-1.3	0.0	0.1	1.2	-0.1	0.3	-0.1	0.0	0.3
100	0.8	-0.1	0.4	-0.5	-0.1	0.3	2.6	-0.2	0.8	1.0	-0.2	0.7
200	2.4	-0.5	1.2	0.7	-0.4	0.9	5.4	-0.8	2.5	3.0	-0.6	2.2
300	4.0	-1.1	2.5	1.7	-0.8	2.0	9.4	-1.9	4.6	5.5	-1.4	4.4
Middle latitudes (O3=325 DU, SZA=40), Actual Profile: 20 km, *a priori*: 18 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	1.1	-0.1	0.3	-0.2	0.0	0.2	2.6	-0.1	0.5	1.3	-0.1	0.5
100	2.1	-0.2	0.6	0.5	-0.1	0.5	4.2	-0.3	1.3	2.5	-0.2	1.1
200	3.9	-0.5	1.8	1.8	-0.4	1.5	7.9	-0.9	3.3	5.1	-0.7	3.1
300	6.1	-1.2	3.6	3.1	-0.8	3.1	14.0	-2.4	5.2	9.2	-1.7	5.1
High latitudes (O3=375 DU, SZA=60), Actual Profile: 20 km, *a priori*: 18 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO2 (DU)	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}	εSO2	εO3	Res_{312}
50	3.6	-0.1	0.5	1.8	-0.1	0.5	5.8	-0.1	0.9	4.2	-0.1	0.9
100	4.9	-0.2	1.3	2.8	-0.2	1.2	8.2	-0.4	2.1	6.1	-0.3	2.0
200	8.1	-0.8	3.3	4.9	-0.5	3.1	15.7	-1.5	4.2	11.7	-1.1	4.2
300	13.2	-1.9	5.2	8.0	-1.2	5.1	34.9	-5.0	5.6	24.5	-3.5	5.6

Figure S1. The above plot shows the differences between Step 2 and Step1 ozone and SO2 for the El Chichon case shown in Figure 5. It can be seen that the O3 errors corrected in Step 2 are anti-correlated with SO2 step 1 errors.

Figure S2. Matrix elements of O_3 (left) and SO_2 (right) plotted for the 317, 331 and 340 channels at positions 3 (dotted) and 18 (solid) along the TOMS cross track (positions range from 1 to 35) for one orbit. These plots represent the sensitivity of O3 and SO2 to a perturbation of 1 N-value.

Figure S3. Mean 340 nm N-value correction with error bars computed from a sample of 90 orbits, shown as a function of the TOMS swath position. The error bars shown are estimated from the standard deviation computed from the 90 orbits. The correction shown is subtracted from the measured N_{340}-value.

Figure S4 – Total ozone maps for June 17 and June 18 following the Mount Pinatubo eruption on June 15, 1991: for June 17, a) MS_SO2 and c) Krueger-Kerr and for June 18 b) MS_SO2 (and d) Krueger-Kerr . In both cases the total ozone for KK shows anomalously low values inside the SO_2 plume. The maps for MS_SO2, which include a Step 2 O_3 correction, show the effects of aerosols on the ozone retrieval at 331 nm.

Figure S5 – Scatterplots comparing MS_SO2 and Krueger-Kerr retrievals on June 17 and June 18, 1991 for a) SO2 and b) Total Ozone.