

5 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 ~ 1.1 nm) 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 $3^\circ \times 3^\circ$ 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.

S3.1 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)

$$\mathbf{K} = \begin{pmatrix} \frac{\partial N_{317}}{\partial \Omega} & \frac{\partial N_{317}}{\partial \Sigma} & \frac{\partial N_{317}}{\partial R} (\lambda_1 - \lambda_R) & \frac{\partial N_{317}}{\partial R} \\ \frac{\partial N_{331}}{\partial \Omega} & \frac{\partial N_{331}}{\partial \Sigma} & \frac{\partial N_{331}}{\partial R} (\lambda_2 - \lambda_R) & \frac{\partial N_{331}}{\partial R} \\ \frac{\partial N_{340}}{\partial \Omega} & \frac{\partial N_{340}}{\partial \Sigma} & \frac{\partial N_{340}}{\partial R} (\lambda_3 - \lambda_R) & \frac{\partial N_{340}}{\partial R} \\ 0 & 0 & 0 & \frac{\partial N_{380}}{\partial R} \end{pmatrix} \quad (\text{S1})$$

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The reflectivity R_s is computed using the measurement I_m and parameters I_0 , T and s_b from the radiance LUTs defined (see Section 3 New MS_SO2 algorithm, Eq. 3),

$$R_s = \frac{(I_m - I_0)}{T + s_b(I_m - I_0)}. \quad (\text{S2})$$

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S3.2 Step 2 Procedure to correct ozone inside SO₂ 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 O₃ 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. (15a,b) to obtain two values, Ω_1 and Ω_2 , at the position of the target pixel, $\varphi_{\Omega 0}$:

$$\Omega_1 = m_1 \varphi_{\Omega 0} + b_1 \quad (\text{S3a})$$

$$\Omega_2 = m_2 \varphi_{\Omega 0} + b_2 \quad (\text{S3b})$$

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 (16,a,b):

$$w_1 = \frac{d_1}{d_1 + d_2} \quad (\text{S4a})$$

$$w_2 = \frac{d_2}{d_1 + d_2}. \quad (\text{S4b})$$

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

$$\Omega_{cor} = w_1 \Omega_1 + w_2 \Omega_2. \quad (\text{S5})$$

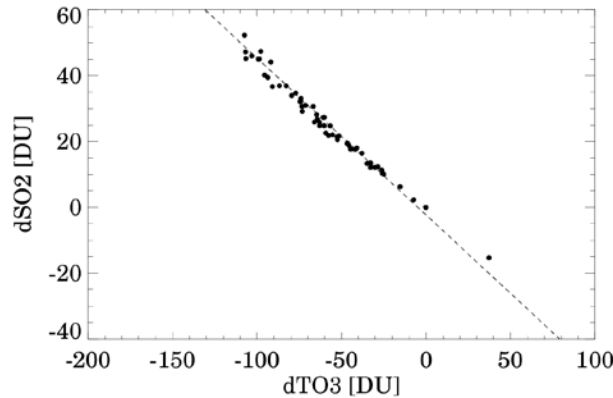


Figure S1. The above plot shows the differences between Step 2 and Step1 ozone and SO₂ for the El Chichon case shown in Figure 5. It can be seen that the O₃ errors corrected in Step 2 are anti-correlated with SO₂ step 1 errors.

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 SO₂ 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 in Eq. (6),

$$G = K^{-1}. \quad (\text{S6})$$

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 O₃ and SO₂ at the three absorbing wavelengths at 317, 331 and 340 nm as a function of latitude. The elements for O₃ and SO₂ 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 O₃, 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.

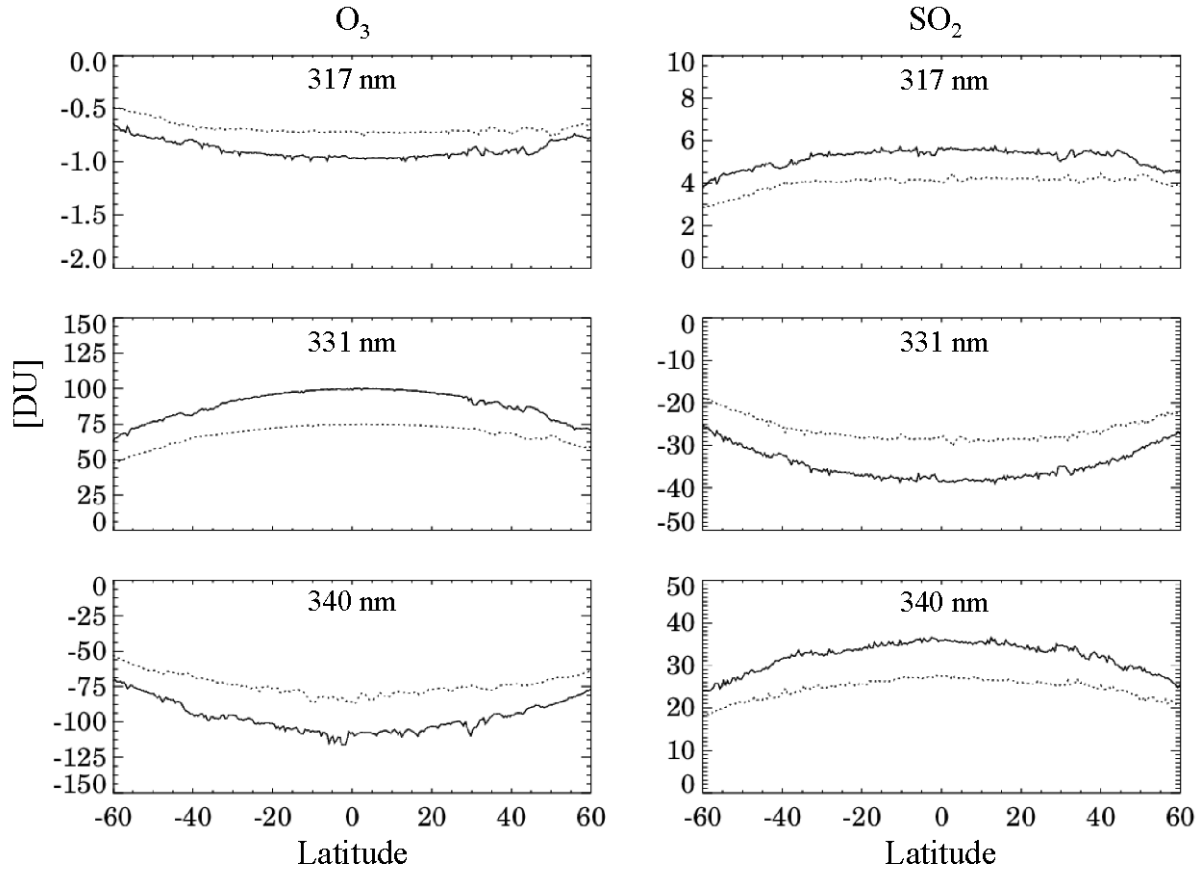


Figure S2. Matrix elements of O₃ (left) and SO₂ (right) plotted for the 317, 331 and 340 channels at positions 3 (dotted) and 18 (solid) along the TOMS cross track (positions range from 1 to 35) for one orbit. These plots represent the sensitivity of O₃ and SO₂ to a radiance perturbation of 1 N-value.

According to Rogers (2000), the error in the retrieval can be defined by the action of G on the measurement error, ε_y , as described by Eq. (S7),

$$dx = G\varepsilon_y. \quad (\text{S7})$$

5 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 O₃ and SO₂ absorption.

With respect to the MS_SO2 algorithm, Eq. (17) can then be expressed as,

$$10 \quad d\Sigma = \frac{\partial\Sigma}{\partial N_{340}} dN_{340}. \quad (\text{S8})$$

where $\partial\Sigma/\partial N_{340}$ and $d\Sigma$ represents the G matrix element for SO₂ at 340 nm and the SO₂ retrieved in the SO₂ free regions of the atmosphere, respectively.

15 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 60°N 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,

$$dN_{340} = \text{Mean}\left(\frac{d\Sigma}{\frac{\partial\Sigma}{\partial N_{340}}}\right) \quad (\text{S8})$$

20 This correction factor was first determined off-line and then applied to the entire data record during operational processing.

S4.2.1 Uncertainties due to SO₂ plume height

Table S1. Percentage error in SO₂ 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°, O₃ = 275 DU).

SO ₂ (DU)	13 km <i>a priori</i> profile*				18-km <i>a priori</i> 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)

25 *Error is calculated as the relative percentage difference in SO₂ 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 SO₂ Jacobians are overestimated (underestimated) with the *a priori* profile and the retrieved SO₂ column amount will be biased low (high).

** Error is calculated as the relative percentage difference in SO₂ 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.

5 **Table S2.** Percentage error in SO₂ 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°, O₃ = 325 DU).

SO ₂ (DU)	13 km <i>a priori</i> profile*				18-km <i>a priori</i> 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.

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Table S3. Percentage error in SO₂ 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°, O₃ = 375 DU).

SO ₂ (DU)	13 km <i>a priori</i> profile*				18-km <i>a priori</i> 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.

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Table S4. Percentage errors in SO₂ (εSO₂) and O₃ (εO₃) total column amounts and *N* value residuals at 312 nm (Res₃₁₂) 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 (O ₃ =275 DU, SZA=10), Actual Profile: 11 km, <i>a priori</i> : 13 km												
SO ₂ (DU)	VZA=0			VZA=60			VZA=0			VZA=60		
	R=0.05	R=0.50	R=0.50	R=0.05	R=0.50	R=0.50	R=0.05	R=0.50	R=0.50	R=0.05	R=0.50	R=0.50
	εSO ₂	εO ₃	Res ₃₁₂	εSO ₂	εO ₃	Res ₃₁₂	εSO ₂	εO ₃	Res ₃₁₂	εSO ₂	εO ₃	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 (O ₃ =325 DU, SZA=40), Actual Profile: 11 km, <i>a priori</i> : 13 km												
	VZA=0			VZA=60			VZA=0			VZA=60		

SO ₂ (DU)	R=0.05			R=0.50			R=0.05			R=0.50		
	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	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

SO ₂ (DU)	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	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 SO₂ (ϵ SO₂) and O₃ (ϵ O₃) total column amounts and *N* value residuals at 312 nm (Res₃₁₂) due to the use of a *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

SO ₂ (DU)	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	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

SO ₂ (DU)	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	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

SO ₂ (DU)	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	Res ₃₁₂	ϵ SO ₂	ϵ O ₃	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.

5 **Table S6.** Percentage errors in SO₂ (ϵ SO₂) and O₃ (ϵ O₃) total column amounts and N value residuals at 312 nm (Res_{312}) due to the use of a *a priori* profile that is 2 km too high (center mass altitude: *a priori*: 18 km, actual: 16 km).

Tropics (O ₃ =275 DU, SZA=10), Actual Profile: 16 km, <i>a priori</i> : 18 km												
SO ₂ (DU)	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	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 (O ₃ =325 DU, SZA=40), Actual Profile: 16 km, <i>a priori</i> : 18 km												
SO ₂ (DU)	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	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 (O ₃ =375 DU, SZA=60), Actual Profile: 16 km, <i>a priori</i> : 18 km												
SO ₂ (DU)	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	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 SO₂ (ϵ SO₂) and O₃ (ϵ O₃) 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).

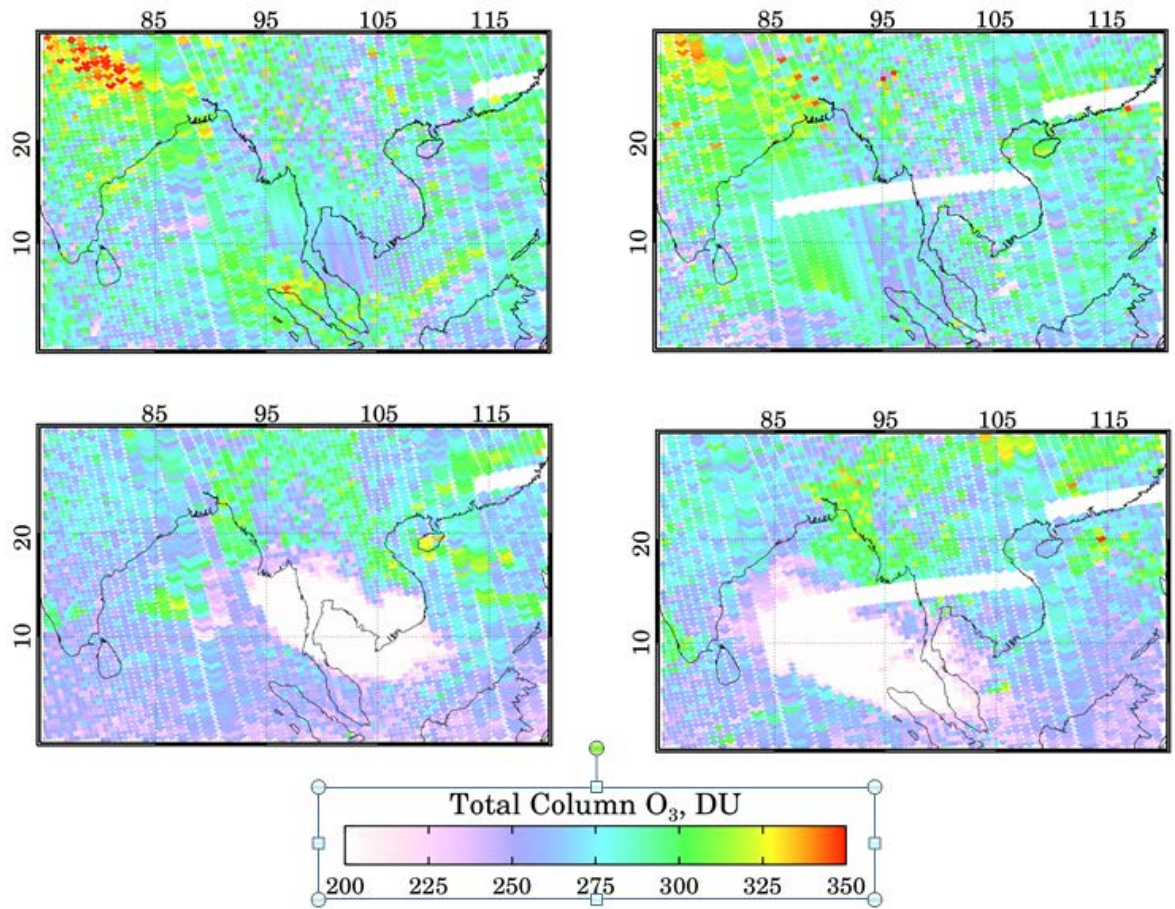
Tropics (O ₃ =275 DU, SZA=10), Actual Profile: 20 km, <i>a priori</i> : 18 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO ₂ (DU)	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}
50	-0.3	0.0	0.1	-1.3	0.0	0.1	1.2	-0.1	0.3	-0.1	0.0	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 (O ₃ =325 DU, SZA=40), Actual Profile: 20 km, <i>a priori</i> : 18 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO ₂ (DU)	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	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 (O ₃ =375 DU, SZA=60), Actual Profile: 20 km, <i>a priori</i> : 18 km												
	VZA=0						VZA=60					
	R=0.05			R=0.50			R=0.05			R=0.50		
SO ₂ (DU)	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	Res_{312}	ϵ SO ₂	ϵ O ₃	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

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S5.1 Mount Pinatubo



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Figure S3 – Total Ozone maps for MS_SO2 (Left) and KK (Right) for June 17 (Top) and June 18 (Bottom) 1991. The total ozone for KK on the right shows anomalously low values inside the SO₂ plume. The maps for MS_SO2 include a Step 2 O₃ correction. The Krueger-Kerr retrieval assumed a radiative transfer AMF.

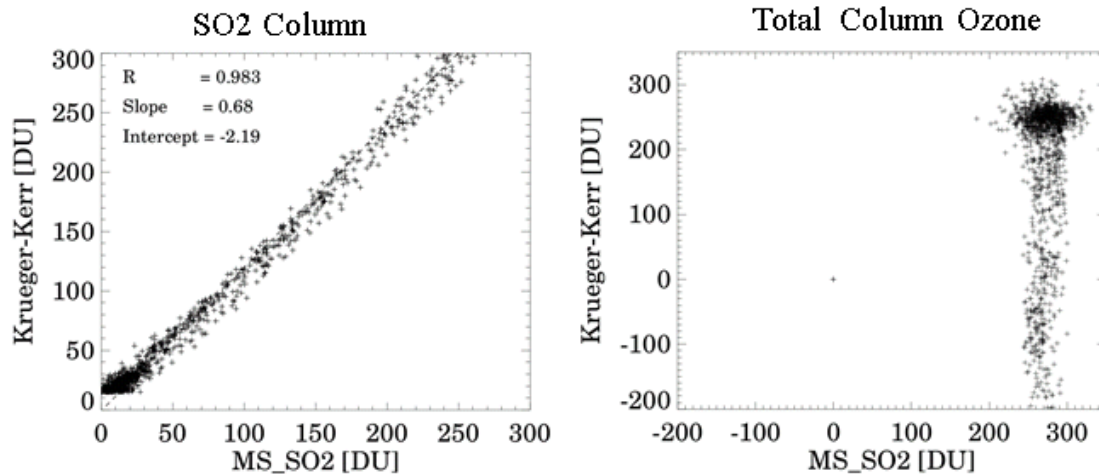


Figure S4 – Scatterplots comparing MS_SO2 and Krueger-Kerr retrievals on June 17 and June 18, 1991 for a) SO2 and b) Total Ozone.