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Ozone profile retrievals from the Ozone Monitoring Instrument

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Abstract

Ozone profiles from the surface to about 60 km are retrieved from Ozone Monitoring Instrument (OMI) ultraviolet radiances using the optimal estimation technique. OMI provides daily ozone profiles for the entire sunlit portion of the earth at a horizontal resolution of 13 km×48 km for the nadir position. The retrieved profiles have sufficient accuracy in the troposphere to see ozone perturbations caused by convection, biomass burning and anthropogenic pollution, and to track their spatiotemporal transport. However, to achieve such accuracy it has been necessary to calibrate OMI radiances carefully (using two days of Aura/Microwave Limb Sounder data taken in the tropics). The retrieved profiles contain ~6–7 degrees of freedom for signal, with 5–7 in the stratosphere and 0–1.5 in the troposphere. Vertical resolution varies from 7–11 km in the stratosphere to 10–14 km in the troposphere. Retrieval precisions range from 1% in the middle stratosphere to 10% in the lower stratosphere and troposphere. Solution errors (i.e., root sum square of precisions and smoothing errors) vary from 1–6% in the middle stratosphere to 6–35% in the troposphere, and are dominated by smoothing errors. Total, stratospheric, and tropospheric ozone columns can be retrieved with solution errors typically in the few Dobson unit range at solar zenith angles less than 80°.

1 Introduction

Total ozone column and ozone profiles have been retrieved since 1970 from about a dozen Backscattered Ultraviolet (BUV) instruments that have flown on NASA and NOAA satellites. These instruments measure at 12 discrete 1 nm wide wavelength bands in the 250–340 nm range, providing vertical information from the ozone density peak (20–25 km) to ~50 km plus the ozone column down to the surface/cloud altitude (Bhartia et al., 1996). Chance et al. (1991, 1997) showed that it may be possible to extend the ozone profile information to lower altitudes, including the troposphere, by using

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high spectral resolution (<0.5 nm) hyperspectral (contiguous in wavelength) data from instruments like the Global Ozone Monitoring Experiment (GOME). Over the years, several groups, have developed physically based retrieval algorithms to retrieve ozone profiles from GOME radiances (Munro et al., 1998; Hoogen et al., 1999; Hasekamp and Landgraf, 2001; van der A et al., 2002; Liu et al., 2005). Meijer et al. (2006) evaluated ozone profiles retrieved from these algorithms and concluded that though ozone profiles can be retrieved from GOME with better information content in the lower stratosphere than that from Solar Backscatter Ultraviolet (SBUV) instruments, they were not able to demonstrate robust determination of tropospheric ozone.

We have found that in order to derive tropospheric ozone profile it is necessary to perform accurate wavelength and radiometric calibrations and to use an accurate forward model. By making these improvements we demonstrated that valuable tropospheric ozone information can be retrieved from GOME (Liu et al., 2005, 2006a, 2006b). In addition, by using the daily National Center for Environmental Prediction (NCEP) re-analysis tropopause fields we were able to estimate the Stratospheric Ozone Column (SOC) and Tropospheric Ozone Column (TOC), in addition to the Total Ozone column (TOZ).

The NASA Earth Observing System (EOS) Aura satellite was launched on 15 July 2004 into a 705-km sun-synchronous polar orbit with a 98.2° inclination and an equator-crossing time (ascending node) of $\sim 13:45$ (Schoeberl et al., 2006). It carries four instruments, including the Ozone Monitoring Instrument (OMI), to make comprehensive measurements of stratospheric and tropospheric composition. OMI is a Dutch-Finnish built nadir-viewing pushbroom UV/visible instrument that measures backscattered radiances in three channels covering the 270–500 nm wavelength range (UV-1: 270–310 nm, UV-2: 310–365 nm, visible: 350–500 nm) at spectral resolution of 0.42–0.63 nm (Levelt et al., 2006). OMI has a very wide field-of-view (114°) with a cross-track swath width of 2600 km. Measurements across the track are binned into 60 positions for the UV-2 and visible channels and into 30 positions for the UV-1 channel (larger bins due to weaker signals). This results in daily global coverage with a spatial resolution of

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13 km \times 24 km (along \times across track) at nadir position for UV-2 and visible channels and 13 km \times 48 km for the UV-1 channel.

Since OMI measurements are similar to GOME, except for the large swath width and much improved spatial resolution, we apply a modified GOME algorithm to OMI data. It should be noted that there is an operational OMI ozone profile algorithm (van Oss et al., 2001) developed at the Royal Netherlands Meteorological Institute. The overall algorithm is similar to our algorithm; it also uses the optimal estimation technique to retrieve ozone profile from radiances in the spectral region 270–330 nm. But it differs significantly from our algorithm in the implementation (e.g., using different radiometric calibration, a priori covariance matrix, radiative transfer model, retrieved variables, vertical grid).

We will present the retrieval algorithm and validation of the retrievals in four papers. The present paper describes the retrieval algorithm and its key characteristics (e.g., vertical resolution, random-noise and smoothing errors). In the second paper, we will validate the retrievals against the Microwave Limb Sounder (MLS) on Aura to demonstrate that stratospheric ozone profiles can be retrieved accurately from OMI, and SOC can be retrieved from OMI with solution errors comparable to or smaller than those from MLS. MLS is the first limb-viewing instrument to provide accurate estimates of SOC, for it is less affected by ice clouds than the visible and IR instruments. In the third paper, we will validate the retrieved ozone profiles and partial ozone columns in the troposphere against ozonesonde observations. Finally, the fourth paper will focus on comparison of TOZ retrievals with those from the two OMI operational TOZ algorithms. We expect our algorithm to perform better at large solar zenith angles since we account for the effects of ozone profiles while the operational TOZ algorithms assume climatological profiles.

The present paper is organized as follows: The retrieval algorithm is described in Sect. 2. Section 3 discusses the retrieval characterization of OMI retrievals. Since the capability to measure boundary layer ozone from satellites is important for future air quality missions, in Sect. 4 we discuss future strategies for improving these retrievals.

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Section 5 compares OMI retrieval characteristics with those of SBUV and GOME retrievals. Section 6 shows examples of OMI retrievals with a focus on tropospheric ozone.

2 OMI ozone profile retrieval algorithm

- 5 Our retrieval algorithm, initially developed for GOME (Liu et al., 2005), is based on the optimal estimation technique (Rodgers, 2000). It simultaneously and iteratively minimizes the differences between observed and simulated radiance spectra and between retrieved (\mathbf{X}) and a priori (\mathbf{X}_a) state vectors, constrained by measurement error covariance matrix (\mathbf{S}_y) and a priori covariance matrix (\mathbf{S}_a). The cost function χ^2 to be minimized can be written:

$$\chi^2 = \left\| \mathbf{S}_y^{-\frac{1}{2}} \{ \mathbf{K}_i (\mathbf{X}_{i+1} - \mathbf{X}_i) - [\mathbf{Y} - \mathbf{R}(\mathbf{X}_i)] \} \right\|_2^2 + \left\| \mathbf{S}_a^{-\frac{1}{2}} (\mathbf{X}_{i+1} - \mathbf{X}_a) \right\|_2^2. \quad (1)$$

- \mathbf{X}_{i+1} and \mathbf{X}_i are the current and previous state vectors, respectively. They consist of ozone column density in a number of atmospheric layers and other auxiliary parameters (which will be described later in this section). \mathbf{Y} is the measurement vector, in our case the logarithm of the sun-normalized radiances. \mathbf{R} is the forward model and the $\mathbf{R}(\mathbf{X}_i)$ are the logarithm of sun-normalized radiances simulated with \mathbf{X}_i . \mathbf{K}_i is the Fréchet derivative or weighting function matrix, defined as $\partial R / \partial \mathbf{X}_i$. The *a posteriori* solution is given as:

$$\mathbf{X}_{i+1} = \mathbf{X}_i + \left(\mathbf{K}_i^T \mathbf{S}_y^{-1} \mathbf{K}_i + \mathbf{S}_a^{-1} \right)^{-1} \left\{ \mathbf{K}_i^T \mathbf{S}_y^{-1} [\mathbf{Y} - \mathbf{R}(\mathbf{X}_i)] - \mathbf{S}_a^{-1} (\mathbf{X}_i - \mathbf{X}_a) \right\}. \quad (2)$$

- 20 The main keys to successful retrievals are accurate calibration and simulation of the measurements. This is especially critical for tropospheric ozone retrievals since separation of stratospheric and tropospheric O₃ requires better than 0.2–0.3% accuracy in measuring and modeling the radiances in the Huggins band (310–340 nm) (Munro et al., 1998).

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- For GOME, we performed several radiometric and wavelength calibrations to deal with calibration issues in the spectral region of our interest (Liu et al., 2005). We derived wavelength-dependent slit widths for convolution of high-resolution spectroscopic data and corrected wavelength-dependent wavelength shifts. We performed an under-sampling correction to the spectral sampling in GOME. Due to the relative intensity offset between GOME band 1 (<312 nm) and GOME band 2 (>312 nm) and different degrees of degradation, we fitted a wavelength-dependent correction in band 1 constrained by the measurements from band 2. Finally, we optimized the fitting windows (290–307 nm, 326–339 nm) and avoided spectral regions with significant calibration problems; the spectral region 307–325 nm is significantly affected by inadequate polarization correction to GOME radiances and variable slit widths, and wavelengths below 290 nm contain large measurement errors. To accurately simulate the measurements, we used the Linearized Discrete Ordinate Radiative Transfer model (LIDORT) (Spurr, 2002) with a look-up table to correct for large radiance errors due to the neglect of polarization in the radiance calculation, and we modeled the Ring effect directly using a single scattering model (Sioris and Evans, 2000). We also improved the forward model inputs of temperature, surface pressure, surface albedo, clouds, and aerosols. With these improvements in GOME calibrations and forward model calculations, the fitting residuals in the Huggins bands were reduced to ~0.2% (Liu et al., 2005), and later to ~0.1% with a further set of improvements (Liu et al., 2007).

- We have adapted our GOME algorithm to process OMI version 3 level 1b data. Since the OMI instrument differs from GOME in many important ways: GOME optics is polarization sensitive, OMI uses a depolarizer; GOME mechanically scans and measures radiances from cross-track pixels using the same detector elements, OMI images the cross-track pixels onto different detector elements; OMI has much wider swath than GOME (Dobber et al., 2006; Levelt et al., 2006), they present somewhat different calibration and forward model challenges. The version of LIDORT used in the GOME algorithm includes a pseudo-spherical correction for the solar beam, but not for the line of sight direction. Failure to account for the Earth's curvature can lead to radiance

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errors of 5–10% for viewing zenith angles in the range 55–70° (Spurr, 2004). In addition, the previously used look-up table for polarization correction is not accurate for large viewing zenith angles. Therefore, we now use the most recent version of Vector LIDORT (VLIDORT) (Spurr, 2006) and use a different approach to perform polarization correction. Modifications with respect to calibration and radiative transfer simulations are described in detail in the following paragraphs.

Because OMI uses a polarization scrambler to depolarize the measurement signal, and the instrumental slit function only exhibits small spectral variation, OMI radiances over 307–325 nm are much easier to model than GOME radiances. Therefore, we choose a different fitting window for OMI, 270–310 nm from UV-1 and 310–330 nm from UV-2. Due to difference in spatial resolutions between UV-1 and UV-2, two UV-2 spectra are co-added to match the UV-1 spatial resolution, and therefore retrievals are done at the UV-1 spatial resolution. To speed up the retrievals and reduce the measurement noise, we co-add 5 adjacent spectral pixels in UV-1 (2 for UV-2). Since there is no significant natural variation in the solar irradiance at the OMI wavelengths, and individual irradiance spectra measured by OMI have both short-term noise and seasonally varying errors, we use the mean solar irradiances derived from three years of OMI data (2005–2007) to normalize the earthshine radiances except that they are adjusted for Sun-Earth distances. To account properly for OMI instrumental slit functions, we derive the slit widths from the solar irradiance spectrum separately for each channel by cross-correlating the OMI irradiance spectrum to a high-resolution solar irradiance reference spectrum (Caspar and Chance, 1997; Chance, 1998), assuming a Gaussian slit function in the convolution. OMI level 1b radiances include in-flight wavelength calibration, so we do not need to correct wavelength registration before the retrievals; we only fit two radiance/irradiance shift parameters (one for each channel) in the retrievals to account for residual wavelength registration errors. Compared to GOME, OMI spectral sampling is significantly improved, especially UV-2 (Chance et al., 2005). The undersampling correction implemented in GOME only has negligible effects on both fitting residuals and retrievals, so this correction is not used in the OMI

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algorithm. The OMI instrument has been very stable through 2008, with degradation of ~2% in UV-1 and ~1% UV-2. We do not fit a wavelength-dependent correction the UV-1 channel to account for varying intensity offsets between the two channels. In fact, doing so reduces the ozone information.

OMI uses a 2-D CCD detector array, where each cross-track position uses a different row of detectors. All the operational level-2 products have shown across-track-dependent biases due to limited pre-launch calibration as a function of detector position. Our preliminary retrievals indicate both wavelength and cross-track position dependent errors in our fitting window. To investigate the quality of OMI radiometric calibration and perform necessary corrections to OMI radiances, we simulate OMI radiances and compare them with the observed radiances. The MLS instrument on the Aura satellite measures ozone profiles down to 215 hPa with vertical resolution of ~3 km. Its ozone products have been extensively validated (Jiang et al., 2007; Froidevaux et al., 2008; Livesey et al., 2008). Thus, MLS provides an excellent source of ozone to check the OMI calibration. In the simulation, we use zonal mean v2.2 MLS ozone profiles for pressure <215 hPa, and climatological ozone profiles from McPeters et al. (2007) for pressure >215 hPa. We examine the average differences between simulated and observed radiances over the tropics, where there is less spatiotemporal variability in ozone.

Figure 1 shows the mean radiance differences in the spectral range 270–350 nm for different UV-1 cross-track positions, derived from clear-sky conditions on 11 July 2006. The overall features do not vary from day to day. The differences typically vary from -6%–7% and are up to ~10% in 300–310 nm for the first and last cross-track positions. There are significant wavelength and cross-track dependences, and there are discontinuities of 3–9% at 310 nm between UV-1 and UV-2. Some of the spikes around 280 and 285 nm are partly due to emission from Mg⁺ and Mg in the ionosphere (Joiner and Aikin, 1996), which is not included in our forward model simulation. There are some high frequency structures between 300–310 nm that cannot be explained either by errors in wavelength calibration or possible errors in MLS ozone profiles used

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in our simulation. The smaller differences at longer wavelengths are related to small errors in our cloud correction algorithm.

Although forward model parameter errors in the simulation (e.g., errors in MLS and climatological ozone) can contribute to some of the differences shown in Fig. 1, the presence of consistent overall features for a number of time periods indicates the existence of calibration problems in OMI level 1b data. The radiances at shorter wavelengths should not depend on cloudiness. However, we find that these differences increase with increasing cloudiness, suggesting the existence of straylight errors in UV-1 and at the shorter UV-2 wavelengths. Since we cannot derive good tropospheric ozone in presence of these errors, we apply a first-order correction to OMI radiances using the average percent difference between measured and simulated radiances derived from 2 days of MLS data in the tropics. This so-called “soft” (also called vicarious) calibration is applied independent of time and latitude, so our retrievals are still affected by residual straylight errors that very likely vary seasonally and latitudinally. These errors are still under investigation.

We use the VLIDORT model to calculate radiances and weighting functions (Spurr, 2006, 2008). The model implements the complete vector discrete ordinate solution with full linearization facility (analytic weighting functions), deals with attenuation of solar and line-of-sight paths in a curved atmosphere, and includes an exact treatment of the single scatter computation. VLIDORT can be run in scalar-mode only (without polarization), which is faster by almost an order of magnitude than a vector calculation. For OMI retrievals, we adopt the following procedure to optimize radiative transfer calculations.

We perform both scalar-only and full-polarization calculations at ~ 10 selected wavelengths and derive polarization corrections at these wavelengths. Next, we perform scalar-only calculations at all other wavelengths, and then interpolate the polarization corrections. The radiance calculation time for the retrieval spectral window is then faster by a factor of ~ 6 than that achieved using VLIDORT in full-polarization mode at all wavelengths; accuracy is maintained to better than 0.1%.

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The radiance calculation is made for a Rayleigh atmosphere (no aerosols) with Lambertian reflectance assumed for the surface and for clouds (treated as reflecting boundaries). For GOME, we used climatological aerosols in the retrievals (Liu et al., 2005). For OMI, we decided to switch this option off because the use of wavelength-dependent surface albedo can partly account for the presence of aerosols just like the use of climatological aerosols. In contrast to the situation with the GOME retrieval algorithm, trace gases other than ozone are not modeled and retrieved. This only slightly affects retrievals except for volcanic eruption conditions. Retrievals of SO_2 and BrO will be added later, since there is adequate spectral information in our fitting window for these trace gases. High-resolution ozone cross sections (Brion et al., 1993), convolved with fitted OMI slit functions and weighted with a high-resolution solar reference spectrum, are used in the simulation to reduce the computation time. For radiative transfer above a reflecting cloud boundary, we take the cloud-top pressure from the OMI $\text{O}_2\text{-O}_2$ algorithm (Acarreta et al., 2004). The monthly mean cloud climatology derived from OMI Raman cloud products (Joiner and Vasilkov, 2006) is used to fill in where cloud-top pressure is not available from the $\text{O}_2\text{-O}_2$ algorithm. An initialized cloud fraction is determined from a single wavelength around 347 nm (degraded to 1.1 nm spectral resolution), based on surface albedo taken from an improved TOMS-derived database (O. Torres, personal communication, 2008). The cloud fraction is then retrieved as an auxiliary parameter. To account for the temperature dependence of ozone absorption, we use daily temperature profiles from NCEP reanalysis data (Kalnay et al., 1996). Due to the much higher spatial resolution of OMI compared with NCEP data, we do not use the NCEP surface pressure; instead, surface pressure is derived from the topographical altitude of the OMI pixel by assuming a standard sea level pressure of 1 atm.

Our state vector contains partial ozone column density (in DU, $1 \text{ DU} = 2.69 \times 10^{16} \text{ molecules cm}^3$) in 24 layers. The 25-level vertical pressure grid is set initially at $P_i = 2^{-i/2} \text{ atm}$ for $i=0, 23$ and with the top of the atmosphere set for P_{24} . This pressure grid is then modified by the surface pressure and daily NCEP thermal tropopause pres-

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sure. Each layer is thus approximately 2.5-km thick, except for the top layer. There are 4 to 7 layers in the troposphere, depending on the tropopause height. There are several different definitions of tropopause, and which tropopause to use for defining TOC is controversial (Liu et al., 2006b; Stajner et al., 2008). We note, however, that the primary purpose of using daily data of tropopause pressure from NCEP is to derive TOC and SOC. For the retrieval of ozone profiles, it is unnecessary to use a tropopause. TOC and SOC can be re-calculated from our retrievals through interpolation if accurate knowledge of the tropopause is available.

In addition to the 24 ozone values, our state vector also contains wavelength-dependent surface albedo (constant surface albedo for UV-1 and first-order polynomial for UV-2), scaling parameters for the Ring effect (1 parameter for each channel), radiance/irradiance wavelength shifts (1 parameter for each channel), wavelength shifts between radiance and ozone cross sections (first-order polynomial for each channel), cloud fraction, and scaling parameters for mean fitting residuals (1 parameter for each channel). To constrain the retrievals, we use climatological mean ozone profiles and their standard deviations derived from 15 yr of ozonesonde and Stratospheric Aerosol and Gas Experiment (SAGE) as a priori, which varies with latitude and month (McPeters et al., 2007). A correlation length of 6 km is used to construct the off-diagonal terms of the a priori covariance matrix. We use OMI random-noise errors from the level 1b data as measurement errors. However, fitting residuals ($\sim 0.45\%$ in UV-1 and $\sim 0.07\%$ in UV-2 on average) for successful retrievals are typically half the size of the measurement random-noise errors. This suggests that those errors in OMI data are overestimated. Due to performance considerations (use of the on-line radiative transfer calculations and the large OMI data volume), we currently limit the retrievals to regions of high scientific interest and pixels collocated with other correlative ground-based or satellite measurements.

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3 Retrieval characterization

The Averaging Kernels (AK) matrix \mathbf{A} , whose i th row \mathbf{A}_{ij} ($j=1, n$, where n is the number of layers) describes about how the retrieved profile in a particular layer i is affected by changes in the true profile \mathbf{X}_T in all layers, characterizes the retrieval sensitivity and vertical resolution of the retrieved profile. Though it can be calculated by perturbation analysis for any type of retrieval algorithm, the optimal estimation retrieval technique provides a closed-form solution for \mathbf{A} (Rodgers, 2000):

$$\mathbf{A} = \frac{\partial \mathbf{X}}{\partial \mathbf{X}_T} = (\mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K} + \mathbf{S}_a^{-1})^{-1} \mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K} = \hat{\mathbf{S}} \mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K} = \mathbf{G} \mathbf{K}, \quad (3)$$

where $\hat{\mathbf{S}}$ is the solution error covariance matrix, and \mathbf{G} is the matrix of contribution functions. The diagonal elements of \mathbf{A} , known as Degrees of Freedom for Signal (DFS), describe the number of useful independent pieces of information available at each layer from the measurements. The trace of \mathbf{A} is the total DFS for the retrieval. Similarly, the sum of the diagonal elements in the troposphere (stratosphere) is the tropospheric (stratospheric) DFS. The AKs for TOZ, SOC, and TOZ can be derived from \mathbf{A} by summing up the rows of \mathbf{A} in all, stratospheric, and tropospheric layers, respectively. To avoid confusion, we will call those AKs as Column Averaging Kernels (CAK) or \mathbf{A}_c . The TOZ CAK represents the fraction of actual ozone columns deviating from the climatology at individual layers that can be retrieved in the entire profile. Therefore, it is essentially the same quantity as the Retrieval Efficiency (RE) (Hudson et al., 1995), an important concept for TOZ retrievals because it indicates what fraction of ozone in the lower troposphere or boundary layer can be captured in the retrieved TOZ. It should be noted that though AK and these derived quantities are mainly determined by the inherent physics (i.e., \mathbf{K}), they do depend on the measurement errors, atmospheric variability, and the correlation length.

Retrieval error is another important quantity characterizing the quality of the retrievals. It consists of random and systematic errors from measurements and forward model simulations, and smoothing errors, which occur because the vertical resolution

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of the retrieved profile is coarser than the thickness of the layer in which the profile is reported (It should be noted that though the use of coarser layers in the state vector would reduce the smoothing errors, it increases the forward model errors. Our choice of ~2.5 km thick layer is a compromise between the two). The random-noise error covariance matrix \mathbf{S}_n and smoothing error covariance matrix \mathbf{S}_s can be directly estimated from the retrievals (Rodgers, 2000):

$$\mathbf{S}_n = \mathbf{G}\mathbf{S}_y\mathbf{G}^T \quad (4)$$

$$\mathbf{S}_s = (\mathbf{A} - \mathbf{I})\mathbf{S}_a(\mathbf{A} - \mathbf{I})^T \quad (5)$$

The sum of \mathbf{S}_n and \mathbf{S}_s is $\hat{\mathbf{S}}$ as seen in Eq. (3). The square root of diagonal elements of the \mathbf{S}_n , \mathbf{S}_s , and $\hat{\mathbf{S}}$ are the random-noise errors (i.e., precisions), smoothing errors, and solution errors, respectively. The solution errors are the root sum square of the random-noise and the smoothing errors. Corresponding errors in TOZ, SOC, and TOC can be easily calculated from the matrices by adding up errors at individual layers and removing correlated errors among different layers, so those integrated errors are usually smaller than the sum of errors at individual layers. Errors contributed from each layer to the overall TOZ, SOC, and TOC smoothing errors, i.e., the Column Error Contribution Function (CECF) E_c , can be derived similar to Eq. (5) as:

$$E_c = (\mathbf{A}_c - \mathbf{I}_c)X_{ae} \quad (6)$$

where X_{ae} is the a priori error, \mathbf{I}_c refers to the idealized CAK. For TOZ, the values of \mathbf{I}_c are 1 at each layer; for SOC/TOC, the values are 1 in the stratosphere/troposphere, but 0 in the troposphere/stratosphere.

Generally, the solution errors are dominated by the smoothing errors. With regard to errors due to forward model and forward model parameter assumptions, we have found from GOME that these errors are generally small compared to the smoothing errors (Liu et al., 2005), so they will not be discussed here. Systematic measurement errors are the most difficult to evaluate; this is largely due to lack of full understanding

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of the OMI instrument calibration. We will determine systematic measurement errors remaining after soft calibration, by means of intercomparison with other correlative measurements.

Figure 2a shows one orbit of retrieved ozone profiles on 11 July 2006. This orbit overpasses the Eastern Pacific Ocean, goes through Alaska and extends to the Arctic sea. Figure 2b shows the fitted effective cloud fraction and zero-order surface albedo in UV-2, and the used effective cloud-top pressure corresponding to the retrievals. Some low/middle level clouds (with effective cloud fraction >0.5) are located around 10° N and 50° N. Surface albedo in the UV is normally 5–8% over the ocean and 2–4% over land (Herman and Celarier, 1997); the fitted albedo shows elevated values at 10° N–40° N due to sun glint, at 70° N–85° N due to sea ice in the Arctic sea, and sometimes over cloudy areas, probably due to inadequate cloud modeling. Ozone number densities are highest in the pressure range 20–75 hPa (20–27 km) depending on the latitude, and closely follow the tropopause (white line). Low ozone in the tropical troposphere is transported to the middle and upper troposphere at Northern middle-latitudes (e.g., 30° N–45° N) with the ridge of subtropical upper tropospheric fronts (Hudson et al., 2003). High tropospheric ozone at 28° N and 50° N is likely caused by the transport of stratospheric ozone in a folding event (mainly located at 50° N but with one of its tongues extending to 28° N; this can be seen clearly in the TOZ map, not shown here).

Figures 3 and 4 show the retrieval characterization corresponding to the retrievals in Fig. 2, including DFS, random-noise errors, smoothing, and solution errors at each layer (the top layer from 0.35 hPa to the top of the atmosphere is not shown due to its broad extent) as well as in TOZ, SOC, and TOC. Profile AKs, CAKs and CECFs of TOZ, SOC, and TOC, for the three clear-sky retrievals indicated as 1, 2, 3 in Figs. 2a and 3a are shown in Figs. 5–7. Because ozone values vary by more than two orders of magnitude at different layers, the AKs in Fig. 5 have been normalized by the actual ozone variability (i.e., a priori error). Table 1 summarizes the average vertical resolution in terms of full width at half maximum and solution errors vs. altitude for three SZA bins (SZA < 30°, 30° < SZA < 60°, 60° < SZA < 80°), and Table 2 summarizes the average total,

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stratospheric and tropospheric DFS and solution errors in TOZ, SOC, and TOC. The overestimate of measurement errors in OMI level 1b data has been corrected in all these figures and tables; this correction increases mean total DFS by 0.75 (0–1.2) and tropospheric DFS by 0.15 (0–0.3) and slightly reduces retrieval errors.

5 Figure 3a shows the DFS at each layer. In the stratosphere, the DFS is generally highest over the pressure range 1–30 hPa (25–45 km). There is a second maximum around the tropopause for about 50° N–80° N. There is little tropospheric information for 60° S–35° S due to limited photon penetration into the troposphere as a result of high SZA and low surface albedo conditions. Under other clear-sky conditions, the
10 DFS generally peaks in the 500–700 hPa layer. Sometimes, DFS can peak in the first layer when there are low-level clouds or snow/ice surfaces. The relatively weak vertical information in the tropical upper troposphere is because SZA is low, ozone column in the stratosphere is small, and multiple scattering generally peaks at lower altitudes. However, the RE for this altitude range is almost 1, since ozone in this altitude range
15 can be well captured in the ozone profile, but will be retrieved at a broad altitude range.

The total DFS value is ~6–7.3, with values of 5–6.7 in the stratosphere and 0–1.5 in the troposphere (Fig. 4a, Table 2). Stratospheric DFS usually increases at high SZA due to the longer photon path length that increases the vertical discrimination of ozone at higher altitudes. Tropospheric DFS decreases quickly at SZA larger than 60°
20 for the same reason (reduced photon penetration to lower altitudes). At 35° S–45° N, tropospheric DFS is not adversely affected by the existence of low-level clouds and is sometimes enhanced, because clouds enhance ozone sensitivity above them, as well as shielding information below them.

From the examples shown in Fig. 5a–c, we can see that AKs are well defined and
25 ozone profiles are well resolved in the stratosphere. The average vertical resolution is about 7–11 km over the pressure range 1.5–100 hPa or 15–45 km (Table 1). In the troposphere, some AKs are not well defined due to inadequate sensitivity (Fig. 5a) and most AK peak altitudes are not coincident with their nominal altitude values. Where the AKs are defined (peak altitudes are within 6 km of nominal altitudes), the average

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vertical resolution is 10–14 km (Table 1).

The REs or TOZ AKs (blue lines in Fig. 6) are generally ~1 above the first layer except for ~60° S–40° S due to little information (e.g., Fig. 6a) or regions with high-level clouds; the oscillations around 1 arise from the assumed a priori covariance matrix.

5 The RE in the first layer (centered at ~850 hPa) is generally ~0.4–0.7 for most of the tropical and mid-latitude summer clear-sky conditions (e.g., Fig. 6b–c), and the corresponding effective photon penetration depth is 800–900 hPa for these conditions. When there are low-level clouds or snow/ice surfaces (e.g., 70° N–80° N), the RE in the first layer can be greater than 0.9.

10 When there is little tropospheric information, the TOC CAK centers around zero and SOC CAK is almost the same as the TOZ CAK (Fig. 6a). Under other conditions, the SOC (green lines) and TOC (red lines) CAKs in Fig. 6b,c peak in the stratosphere and troposphere, respectively, as expected. But both show significant sensitivity from the other part of the atmosphere, especially in the lower stratosphere and upper troposphere, and both show larger stratospheric oscillations than the TOZ CAK. This seems
15 to indicate large smoothing errors in the retrieved SOC and TOC and the difficulty in separating TOC from SOC. However, it should be noted that the smoothing process operates on the difference in layer ozone column amount (in DU) between actual and a priori ozone profiles (as indicated by the a priori error in Fig. 7) instead of the actual profile itself. Figure 7 shows that the errors contributed from each layer arising from imperfect CAKs are very small in the middle and upper stratosphere (<0.5 DU) and are generally within 2 DU in the lower stratosphere and troposphere. If we assume that errors at individual layers are random an uncorrelated, then the errors in TOZ, SOC, and TOC are the root sum squares of the errors at individual layers. Those integrated
20 errors are very small, 1.4 DU, 2.3 DU, 2.3 DU in TOZ, SOC, and TOC, respectively for Fig. 7b. The positive correlation between adjacent layers in the assumed a priori covariance matrix may slightly reduce those integrated errors; the smoothing errors in TOZ, SOC, and TOC are 1.6, 2.1, and 1.8 DU, respectively for Fig. 7b.

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Figure 3b–d shows the random-noise, smoothing, and solution errors at each layer. The random-noise errors are typically 0.5–2% in the major part of the stratosphere (0.4–76 hPa). They increase to as much as 10% in the lower stratosphere and troposphere (and occasionally to ~16%, e.g., in the upper troposphere and lower stratosphere for 60° S–50° S due to low ozone information). The smoothing errors are generally much larger than random-noise errors, dominating the solution errors, especially over altitude regions with weak ozone information. The solution errors (also shown in Table 1) are typically within 1–6% in the middle and upper stratosphere (1–50 hPa), increasing to 10% (occasionally to 17%) for pressure <1 hPa. In the lower stratosphere and troposphere, they are generally within 6–35% but sometimes as high as 50% for 60° S–50° S due to low ozone information and relatively large climatological variability; the average errors are 8–25% as shown in Table 1.

Figure 4b–d shows corresponding random-noise, smoothing, and solution errors in TOZ, SOC, and TOC. Although these errors vary with many factors including SZA, TOZ, the vertical distribution of ozone, cloud, and surface albedo, the overall errors are quite small except for high SZA > 80°, where the errors increase quickly with the increase of high solar zenith angles. At SZA < 80°, the random-noise errors are within 2 DU for TOZ and SOC and within 3 DU for TOC; the solution errors are within 3.5 DU for TOZ and SOC and within 5 DU for TOC. As shown in Table 2, the average solution errors are 1–2 DU in TOZ, 2–3 DU in SOC, and 2–5 DU in TOC for different SZA ranges. Note that those very small errors (<0.5 DU) in TOZ at 70° N–80° N are due to highly reflecting snow/ice surfaces.

4 Retrieval interferences and further algorithm improvements

The previous section shows that current OMI retrievals effectively exhibit full sensitivity to ozone down to the 800–900 hPa range or the upper part of the boundary layer. It should be noted that sensitivity to boundary layer ozone has not been fully exploited from OMI measurements due to retrieval interferences with other ancillary parameters,

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especially the wavelength-dependent surface albedo parameters. In our retrievals, OMI radiances are inadequately calibrated. In addition, aerosols, clouds, and surface pressure are either not accurately known or are not modeled in the retrievals. So we use the wavelength-dependent surface albedo terms as tuning parameters to account for these effects. However, the limited ozone information for the boundary layer partly originates from the broad variation of ozone absorption with wavelength, which in turn correlates with signatures from aerosols and surface albedo. Thus, fitted wavelength-dependent surface albedo parameters are cross-correlated with ozone parameters, reducing the sensitivity to ozone.

The thin lines in Fig. 4a show the DFS values without interferences from other non-ozone parameters. These values would increase by 0.4–0.7 at 40° S–80° N and the tropospheric DFS would increase by 0.2–0.6, mainly from the first layer (0.1–0.35 from the first layer). For most retrievals at 30° S–80° N, DFS values in the first layer would be comparable or larger than those in the second layer. AKs in the troposphere would be better defined and the vertical resolution would be improved.

Figure 5d–f shows the same AKs as those in Fig. 5a–c except without interferences. We can see significant improvement in the first layer and in the troposphere overall for the second and third examples. The vertical resolution in the troposphere would be improved from 10–14 km to 5–12 km; the RE (thin lines in Fig. 6) would increase to 0.7–0.8 for the first layer, and the effective photon penetration depth would be 920–950 hPa, almost down to the surface. In addition, the oscillations in TOZ, SOC and TOC AKs would be slightly reduced, and retrieval errors would also be reduced.

The effects of interferences suggest that in order to further improve retrievals, especially those in the boundary layer, we need 1) to obtain better instrument calibration, 2) to use other auxiliary information as accurately as possible, and 3) improve the accuracy of the forward model parameters. Further improvements of the retrieval algorithm for OMI could include the addition of longer wavelengths to derive aerosol information, the simulation of radiances at a higher spectral resolution before convolution with instrumental slit functions, the modeling of the bi-directional reflectance distribution

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functions for the surface (including sun-glint), and the treatment of clouds as scattering layers as opposed to Lambertian reflecting boundaries.

Even if we can significantly reduce the retrieval interferences and improve the sensitivity to boundary layer ozone, the lack of adequate vertical information cannot sufficiently separate boundary layer ozone from free tropospheric ozone from UV radiance measurements alone. Combining UV radiance measurements with polarization measurements in the UV (Hasekamp and Landgraf, 2002), and radiance measurements in the Chappuis bands (Chance et al., 1997) and thermal IR (Worden et al., 2007), are likely necessary to improve ozone retrievals in the boundary layer or at the surface. The keys to these combined retrievals are to calibrate different measurements in a consistent manner and to establish spectroscopic databases that are relatively consistent among different spectral regions.

5 Comparison of retrieval characteristics between OMI, GOME, and SBUV

Because ozone profiles have been previously measured from SBUV-like (i.e. from BUUV) and GOME measurement since 1970 and 1995, respectively, it is important to understand the differences in retrieval characteristics (e.g., vertical resolution and retrieval errors) among these measurements. To minimize the effects of factors such as a priori covariance matrix, viewing geometry, ozone fields, and retrieval parameters on the comparison, we modify the OMI level 1b data to SBUV and GOME-like measurements and perform retrievals from the same orbit in Fig. 2. To represent SBUV retrievals, we convolve OMI measurements to the SBUV spectral resolution (1.13-nm FWHM) and interpolate convolved data to SBUV wavelengths (except for 255.5 nm, not measured in OMI). A measurement error of 1% is assumed at each wavelength following the SBUV operational algorithm (Bhartia et al., 1996); the use of 1% error is to account for the scene change during the course of a sequential scan of all the wavelengths. To represent GOME retrievals, we use OMI data in the same spectral regions as our previous GOME retrieval algorithm (290–307 nm, 325–340 nm). We use the same a priori

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covariance matrix and retrieval parameters for all these three retrievals. Tables 3 and 4 show similar comparisons as Tables 1 and 2 but for these three retrievals at SZA between 30–60°.

For GOME retrievals, the total DFS is smaller by 1.7 mainly in the stratosphere because of not using measurements below 290 nm. Corresponding, vertical resolution is significantly coarser than OMI's resolution in the stratosphere and upper stratosphere (13–14 km over 3–100 hPa). The tropospheric DFS is only slightly smaller. The solution errors are larger by 0.5–2% (from 1–6% to 2–8%) at each layer and are slightly larger for TOZ, SOC, and SOC.

For SBUV retrievals, most of the tropospheric DFS (<0.5) is lost and the stratospheric DFS is reduced by 2 due to the use of only 11 discrete wavelengths and a large measurement error. The vertical resolution is 10–14 km over the pressure range 1.5–26 hPa (25–45 km). The vertical resolution in the lower stratosphere and troposphere is 20–25 km, confirming the fact that ozone column below 25 km can still be well derived from the SBUV measurements (Bhartia et al., 1996). The solution errors increase by 1–3% (from 1–6% to 3–7%) in the stratosphere and by 5–10% in the troposphere; errors in TOZ, SOC, and TOZ are more than doubled compared to OMI retrievals. Note that the vertical resolution of 10–14 km above ozone density peak is significantly larger than estimated from the operational algorithm (6–8 km). This is primarily due to the use of a different a priori covariance matrix; an a priori error of 50% is used at each layer to better capture the ozone trend in the operational algorithm, which increases the stratospheric DFS by 1.5.

6 Examples of retrievals

Figure 8 shows global maps of TOZ, SOC, TOC, and cloud fraction on 26 August 2006. Figure 9 shows longitudinal cross sections of ozone below 100 hPa at 10° S and 35.5° N, interpolated to fine vertical grids and converted to volume mixing ratio. Large values of TOZ and SOC at middle latitudes generally correspond to regions of

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tropopause folding, i.e., with large tropopause pressure (white contours on TOZ and SOC maps). TOC in the tropics shows typical wave-1 pattern, with low TOC over regions of strong convection (e.g., the Pacific Ocean) and high ozone over the South Atlantic due to complex coupling between biomass burning, lightning NO_x , and dynamic transport processes (Thompson et al., 2000; Martin et al., 2002; Edwards et al., 2003; Sauvage et al., 2006, 2007). The longitudinal cross section of ozone profiles at 10.5°S in Fig. 9a shows enhanced ozone of 60–90 ppbv in the middle troposphere of South Atlantic, also moderately high ozone of 60 ppbv around Indonesia due to the early stage of the 2006 El Niño event (Logan et al., 2008), as well as low ozone of 20–40 ppbv throughout the troposphere of the Pacific Ocean.

Zonal bands of high TOC are found at middle latitudes (25°N – 55°N , 20°S – 40°S) in both hemispheres. Particularly in the northern middle latitudes, enhanced TOC regions are located over central and eastern US and its outflow area, the west coast of Europe, the Mediterranean, the middle East, East Asia and its outflow regions. These high ozone features are not caused by retrieval artifacts associated with clouds since they do not always correspond to high cloudiness (Fig. 8d). Some of these ozone enhancements will reflect the transport of industrial pollution from the continents; this has been shown from many modeling studies (Parrish et al., 1993; Lelieveld et al., 2002; Liu et al., 2003; Duncan and Bey, 2004; Auvray and Bey, 2005; Li et al., 2005; Cooper et al., 2007). Some of these features could also be caused by stratospheric intrusions (Cooper et al., 2004a, 2005). For example, Fig. 9b shows the transport of high ozone from the stratosphere to the middle troposphere at 150°W , associated with a tropopause folding as seen from the NCEP tropopause. Due to limited vertical resolution and the fact that pollution plumes from continental outflows often mix with stratospheric air masses (Cooper et al., 2004a, 2005), it is difficult to identify the origins of these high ozone features from OMI retrievals alone. It is necessary to use other in-situ observations, model simulations, and meteorological fields to assist with the interpretation of OMI retrievals. It is likely that high ozone in the southern middle latitudes is due to stratospheric intrusion as well as the lifting of ozone precursors from

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biomass burning to the upper troposphere.

There is large spatial variability at middle latitudes, with a mixture of high ozone features with low ozone features due in part to frequent transport of tropical marine air to middle and high latitudes as can be seen from Fig. 8c and Fig. 9b (e.g., 180°W , 140°W , 50°W , 40°E , 140°E , 160°E). Figure 9b shows the transport of low ozone tropical air to the upper troposphere at 120°W .

TOC at high latitudes is generally low partly due to lower tropopause except over Antarctica, where high TOC is because the NCEP tropopause is too high ($<150 \text{ hPa}$). The low TOC over the Himalayas, Greenland, Andes, and Rocky mountains is due to high terrain; when converted to mean mixing ratio, the values are similar to or slightly higher than those in surrounding areas.

The high spatial resolution and daily global coverage of OMI observations make our retrievals especially suitable for tracking the spatiotemporal evolution of ozone features caused by chemical and dynamic processes, such as the long-range transport of pollution, stratospheric folding events, and transport of low-ozone tropical marine air to higher latitudes. Figure 10 shows mean tropospheric ozone mixing ratio as well as longitudinal cross sections of ozone profiles below 100 hPa at 31.5°N and 41.5°N over the North Pacific during an event of transpacific transport of pollution on 5–9 May 2006. This event has been well studied by Zhang et al. (2008); the Asia pollution plume is lifted by a southeastward moving front and is rapidly transported in westerly winds across the Pacific.

OMI retrievals (Fig. 10a) clearly illustrate the progression of this transport event, consistent with Atmospheric Infrared Sounder (AIRS) observations and GEOS-Chem simulations of CO (Fig. 7 of Zhang et al., 2008). The high-ozone stream over the west coast of the US that does not associate with high AIRS CO is not caused by the transport of Asian pollution but by a stratospheric folding event, the spatiotemporal evolution of which can also be clearly seen from Fig. 10a,b. The cross sections also clearly indicate significant stratospheric influences even in regions of transport, although the retrievals are likely overestimated due to smoothing errors that in turn arise from in-

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adequate vertical resolution. The coexistence of stratospheric intrusion with transport of pollution is consistent with the findings of Cooper et al. (2004a, 2004b) that stratospheric air masses often mix with pollution plumes in regions of continental outflow. In the above example, on 9 May, part of the pollution plume reached the west coast of the US. Aircraft observations near 138° W, 42° N show ozone of 50 ppbv in the lower troposphere (0–2.5 km) and 60–80 ppbv (3.5–10 km) in the middle troposphere, and illustrate the decomposition of peroxyacetylnitrate (PAN) and the production of ozone (Fig. 9 of Zhang et al., 2008). OMI ozone profiles over this region are quite consistent with aircraft observations of ozone (Fig. 10c). Another distinct feature from this event is the transport of low-ozone tropical air with the southeastward moving front. Despite coarse vertical resolution, OMI retrievals clearly track horizontal, vertical, and temporal transport of these features. Sometimes low ozone is transported to the upper troposphere, above relatively higher ozone.

7 Summary

We have applied our ozone profile retrieval algorithm, originally developed for the Global Ozone Monitoring Experiment (GOME), to the Ozone Monitoring Instrument (OMI) data. Because OMI instrument characteristics are dissimilar, we use a different strategy to deal with radiometric calibration. The OMI retrievals also use an improved radiative transfer model. To check the radiometric calibration of OMI, we compare radiances simulated with zonal mean Microwave Limb Sounder (MLS) ozone profiles in the tropics with OMI observations. OMI UV radiances show significant across-track and wavelength dependent biases (typically –6–7%) as well as discontinuities of 3–9% at 310 nm between UV-1 and UV-2 channels. A first-order correction is derived by averaging two days' radiance differences, and applied independent of space and time to OMI radiances prior to the retrievals. From corrected OMI radiances (270–330 nm), we retrieve partial ozone columns at 24 approximately 2.5-km layers between the surface and ~60 km using the optimal estimation technique. Total, stratospheric, and

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tropospheric ozone column (TOZ, SOC, TOC) are directly integrated from the retrieved profiles.

There are 6–7.3 degrees of freedom for signal in the retrievals with 5–6.7 in the stratosphere and up to 1.5 in the troposphere. In the stratosphere, ozone information generally peaks between 1–30 hPa with vertical resolution of 7–11 km. In the tropics and mid-latitude summer, tropospheric information generally peaks between 500–700 hPa with vertical resolution of 10–14 km, and the retrievals are effectively sensitive to ozone down to ~800–900 hPa. The random-noise errors (i.e., precisions) are typically 0.5–2% in the middle stratosphere, 3–5% in the upper stratosphere and increase to as much as 10% in the lower stratosphere and troposphere. The solution errors, i.e., root sum square of both random-noise and smoothing errors, are dominated by smoothing errors; they are generally 1–6% in the middle stratosphere, up to 10% in the upper stratosphere, and 6–35% in the troposphere. TOZ, SOC, and TOC can be accurately retrieved. Under solar zenith angles less than 80°, the precisions are generally within 2–3 DU, and the solution errors are within 3–5 DU.

We present several examples of retrievals with an emphasis on tropospheric ozone, although there is much more information in the stratosphere than in the troposphere. OMI retrievals are capable of capturing tropospheric ozone signals due to convection, biomass burning, anthropogenic pollution, transport of pollution, transport of low ozone tropical air to the middle and upper troposphere of middle and high latitudes, and stratospheric intrusions. Despite coarse vertical resolution, OMI's high spatial resolution and daily global coverage make our retrievals suitable for tracking the spatiotemporal evolutions of tropospheric ozone features caused by chemical and dynamic processes.

Due to the vertical distribution of ozone information spanning both the stratosphere and the troposphere, high retrieval precision, accurate estimates of TOZ, SOC and TOC, and OMI's spatial resolution and coverage, OMI ozone profiles constitute a unique and useful dataset to study the distribution of ozone in the troposphere and the stratosphere. This dataset complements ozone measurements from the other three instruments on the Aura satellite: High Resolution Dynamics Limb Sounder (HIRDLS)

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and MLS measure ozone profiles at higher vertical resolutions of 1 and 3 km, respectively, but only in the stratosphere and upper troposphere and with limited spatial coverage; TES focuses on measuring tropospheric ozone with up to two pieces of information in the troposphere, but also with limited spatial coverage.

5 Retrieving ozone in the boundary layer or even at the surface is of great interest for air quality monitoring. However, due to interferences from other retrieval parameters, we cannot yet retrieve all the boundary layer ozone information available in OMI radiance spectra. To further improve the retrievals, we need to continue the improvement of instrumental calibration and perform more accurate radiative transfer simulations. Due
10 to the extensive on-line radiative transfer calculations and large volume of OMI data, it is currently challenging to make retrievals available over the entire OMI period. We will continue to optimize radiative transfer calculations and use more computer resources to speed up the retrievals and make the OMI data record available to the scientific community in the near future.

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Table 1. Average vertical resolution in terms of Full Width at Half Maximum (FWHM) and relative solution errors at different altitudes for three solar zenith angles bins (all-sky conditions) for the orbit of retrievals in Fig. 2. The solution errors are defined relative to the a priori profiles.

| Altitude (km)/ ~pressure (hPa) | FWHM (km) | | | Solution errors (%) | | |
|-----------------------------------|-----------|--------|--------|---------------------|--------|--------|
| | <30° | 30–60° | 60–80° | <30° | 30–60° | 60–80° |
| 52.5/0.6 | 11.8 | 12.8 | 12.1 | 4.6 | 5.8 | 9.5 |
| 47.5/1.2 | 17.5 | 14.5 | 11.3 | 3.4 | 3.8 | 5.0 |
| 42.5/2.3 | 9.6 | 9.5 | 8.9 | 1.9 | 2.0 | 2.7 |
| 37.5/4.5 | 7.7 | 7.6 | 7.5 | 1.2 | 1.4 | 1.7 |
| 32.5/9.0 | 7.2 | 7.1 | 7.3 | 1.1 | 1.6 | 2.1 |
| 27.5/19.0 | 7.6 | 7.8 | 7.6 | 1.3 | 2.0 | 2.6 |
| 22.5/40.2 | 10.7 | 14.3 | 8.3 | 2.5 | 3.4 | 3.4 |
| 17.5/88.0 | 11.6 | 10.7 | 10.9 | 9.6 | 8.3 | 8.1 |
| 12.5/192.3 | 14.3 | 14.1 | 13.9 | 24.7 | 15.6 | 13.8 |
| 7.5/397.0 | 12.5 | 11.4 | 13.7 | 15.2 | 14.5 | 23.3 |
| 2.5/758.0 | 10.0 | 9.6 | 12.2 | 19.7 | 17.2 | 16.7 |

22725

Table 2. Average total, stratospheric, and tropospheric Degree of Freedom for Signal (DFS) and solution errors in total, stratospheric, and tropospheric ozone columns for three solar zenith angles bins (all-sky conditions) for the orbit of retrievals in Fig. 2.

| | DFS | | | Solution errors (DU) | | |
|---------------|------|--------|--------|----------------------|--------|--------|
| | <30° | 30–60° | 60–80° | <30° | 30–60° | 60–80° |
| Total | 6.4 | 6.6 | 6.6 | 1.3 | 1.2 | 2.0 |
| Stratospheric | 5.1 | 5.6 | 6.2 | 2.3 | 2.2 | 2.9 |
| Tropospheric | 1.3 | 1.0 | 0.4 | 1.9 | 2.5 | 4.4 |

22726

Table 3. Similar to Table 1, but for comparison of OMI, GOME, and SBUV retrievals at solar zenith angles 30–60°.

| Altitude (km)/ ~pressure (hPa) | FWHM (km) | | | Solution errors (%) | | |
|-----------------------------------|-----------|------|------|---------------------|------|------|
| | OMI | GOME | SBUV | OMI | GOME | SBUV |
| 52.5/0.6 | 12.8 | 21.5 | 14.1 | 5.8 | 7.7 | 6.6 |
| 47.5/1.2 | 14.5 | 20.1 | 15.7 | 3.8 | 5.2 | 4.9 |
| 42.5/2.3 | 9.5 | 16.2 | 13.8 | 2.0 | 3.4 | 3.6 |
| 37.5/4.5 | 7.6 | 13.5 | 10.7 | 1.4 | 2.3 | 2.9 |
| 32.5/9.0 | 7.1 | 9.4 | 9.9 | 1.6 | 2.2 | 3.8 |
| 27.5/19.0 | 7.8 | 8.9 | 11.4 | 2.0 | 2.4 | 4.1 |
| 22.5/40.2 | 14.3 | 13.3 | 22.6 | 3.4 | 4.0 | 6.0 |
| 17.5/88.0 | 10.7 | 12.6 | 20.4 | 8.3 | 9.7 | 13.2 |
| 12.5/192.3 | 14.1 | 16.7 | 25.0 | 15.6 | 16.9 | 21.5 |
| 7.5/397.0 | 11.4 | 13.4 | 24.6 | 14.5 | 16.4 | 24.8 |
| 2.5/758.0 | 9.6 | 10.8 | 19.2 | 17.2 | 19.1 | 24.9 |

22727

Table 4. Similar to Table 2, but for comparison of OMI, GOME, and SBUV retrievals at solar zenith angles 30–60°.

| | DFS | | | Solution errors (DU) | | |
|---------------|-----|------|------|----------------------|------|------|
| | OMI | GOME | SBUV | OMI | GOME | SBUV |
| Total | 6.6 | 4.8 | 3.7 | 1.2 | 1.5 | 4.1 |
| Stratospheric | 5.6 | 3.9 | 3.5 | 2.2 | 2.7 | 4.6 |
| Tropospheric | 1.0 | 0.9 | 0.3 | 2.5 | 3.3 | 6.3 |

22728

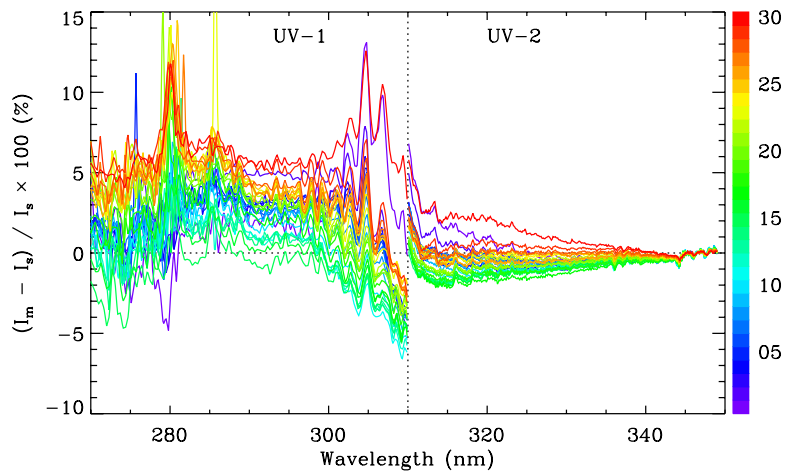


Fig. 1. Mean percentage difference (residuals) between OMI measured (I_m) radiances and those simulated (I_s) using MLS O_3 profiles are plotted as a function of wavelength. The 30 UV-1 across-track positions are shown in different colors. These residuals are derived from nearly clear-sky conditions (effective cloud fraction <20%) in the latitude range 0–20° S on 11 July 2006. Pixels affected by aerosols (OMTO3 aerosol index greater than 1 or less than –1) and sun glint are not used.

22729

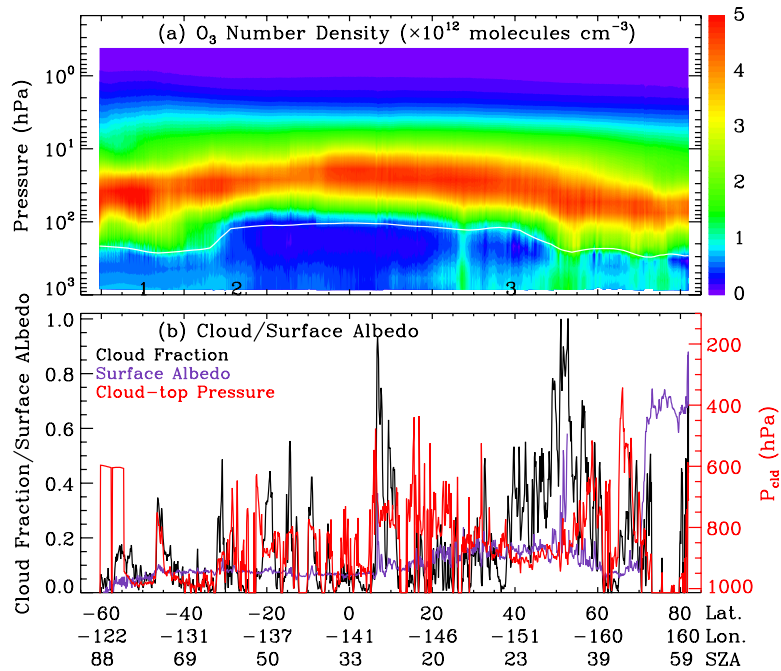


Fig. 2. An orbit of retrievals at across-track position 16 (in the UV-1 channel) on 11 July 2006 as a function of latitude, longitude, and solar zenith angle. **(a)** Ozone profiles in number density, and **(b)** the effective cloud fraction (black), fitted surface albedo (purple) for the UV-2 channel, and effective cloud-top pressure (red) used in the retrievals. The white line in (a) indicates the NCEP thermal tropopause.

22730

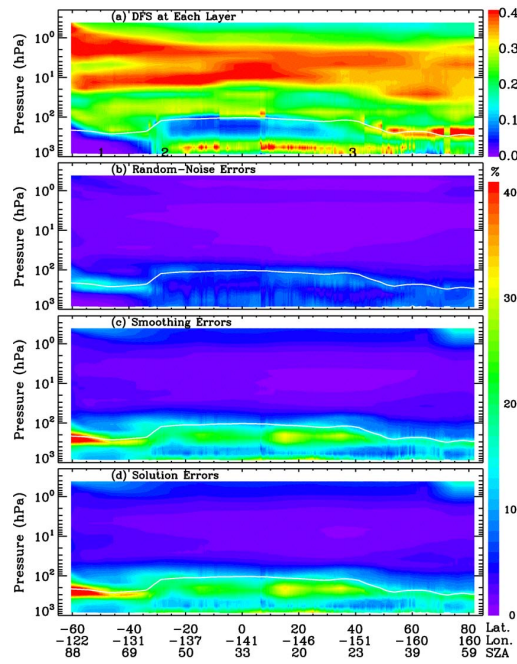


Fig. 3. (a) Degrees of freedom for signal at each layer, (b) 1σ random-noise errors, (c) 1σ smoothing errors, and (d) 1σ solution errors (i.e., root sum square of (b) and (c)) for the same orbit of retrievals as in Fig. 2.

22731

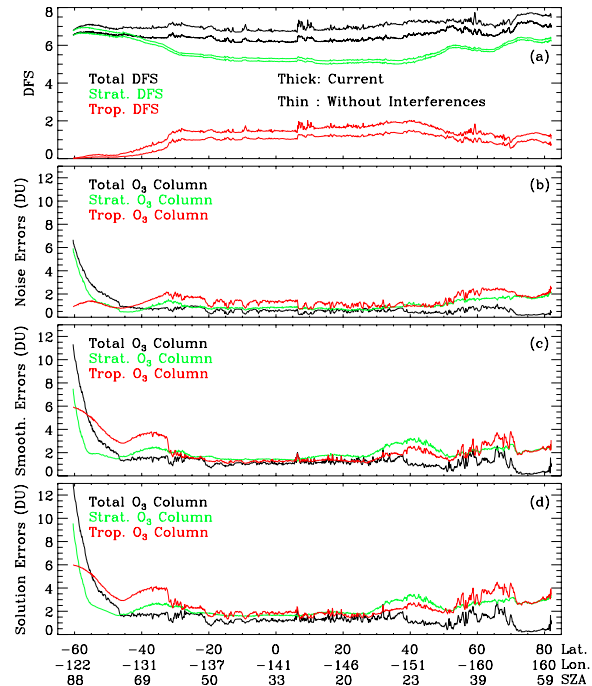


Fig. 4. (a) Total, stratospheric, and tropospheric degrees of freedom for signal, (b) 1σ random-noise errors in total, stratospheric, and tropospheric ozone columns, (c) same as (b) but for 1σ smoothing errors, and (d) same as (b) but for 1σ solution errors (i.e., root sum square of (b) and (c)), for the same orbit of retrievals as in Fig. 2. The thin lines in (a) are for retrievals under ideal conditions (i.e., without interferences from other parameters).

22732

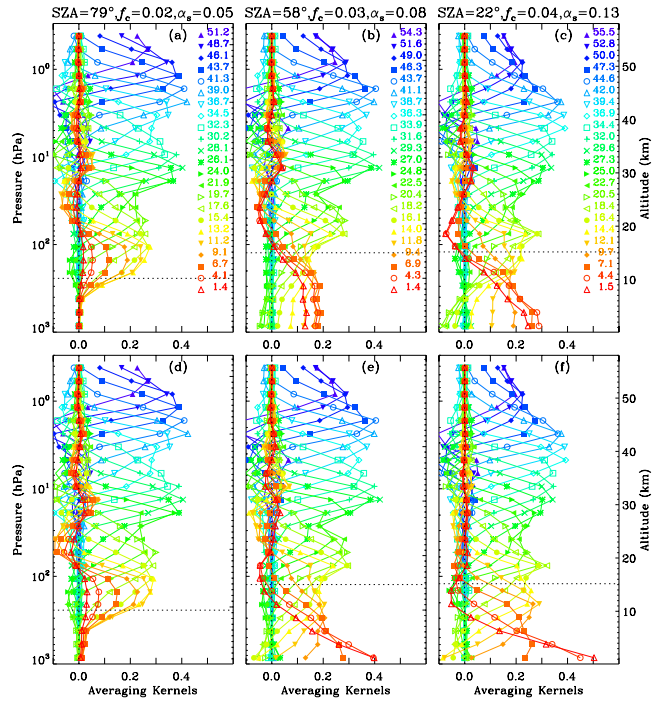


Fig. 5. (a–c) are three examples of nearly clear-sky retrieval averaging kernels for pixels (1)–(3) indicated on Figs. 2a and 3a. (d–e) are the same as (a–c) but under ideal conditions (i.e., without interferences from other parameters). These AKs have been normalized by the actual ozone variability (i.e., a priori error) because ozone values vary by more than two orders of magnitude at different layers. The dotted black horizontal lines indicate the tropopause. The caption on top of each of panel shows solar zenith angle, cloud fraction, and surface albedo.

22733

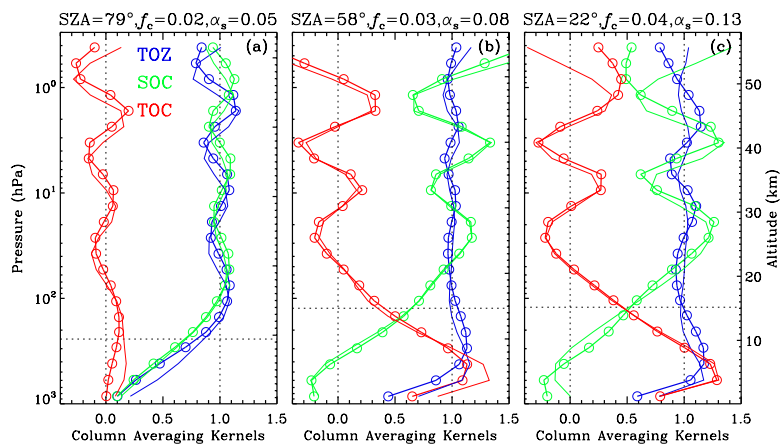


Fig. 6. Column averaging kernels in total, stratospheric and tropospheric ozone columns (TOZ, SOC, and TOC) corresponding to the three examples in Fig. 5. The thick lines with open circles are for current retrievals and the thin lines are for retrieval without interferences.

22734

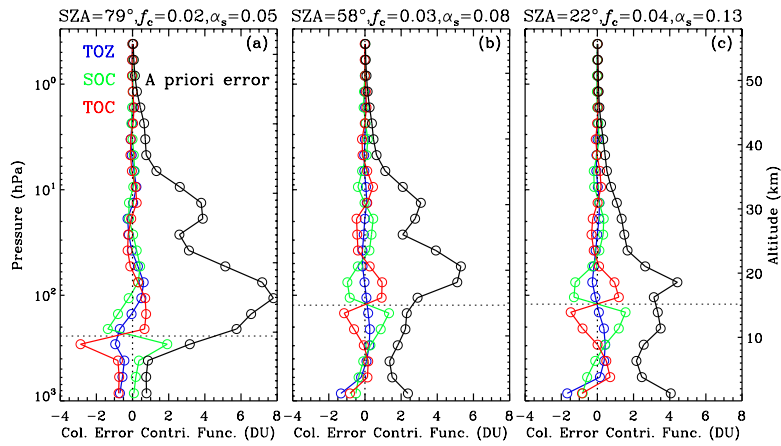


Fig. 7. Column error contribution function (1σ) for total, stratospheric and tropospheric ozone columns (TOZ, SOC, and TOC) corresponding to the three examples in Fig. 5. Also plotted is the a priori error (black) at each layer.

22735

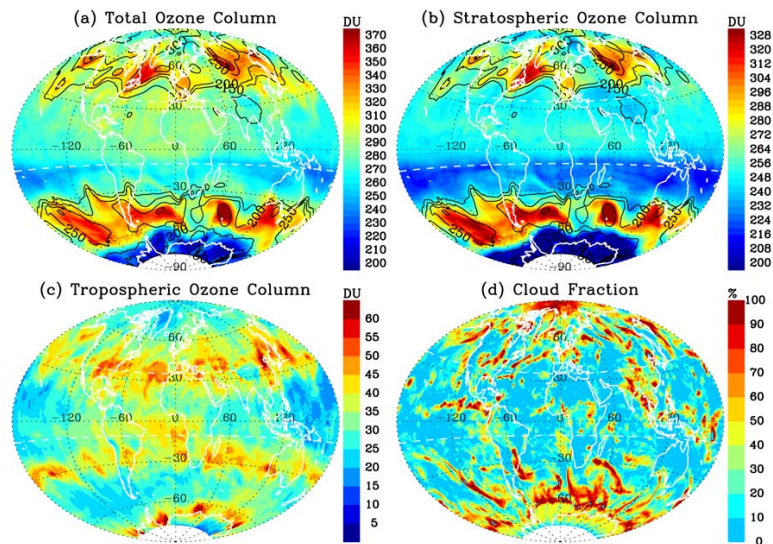


Fig. 8. Maps of total ozone column (a), stratospheric ozone column (b), tropospheric ozone column (c), and cloud fraction (d) on 26 August 2006. The data are mapped on 1° longitude \times 1° latitude grid cells. Some systematic cross-track position dependent biases have been removed before gridding by assuming these data do not vary with cross-track position within a month. The black contours on (a) and (b) indicate the NCEP tropopause pressure used (the contour levels are 100, 150, 200, 250, 300, 350 hPa) and the two white dashed lines indicate the latitudes where zonal cross sections of tropospheric ozone profiles are shown in Fig. 9.

22736

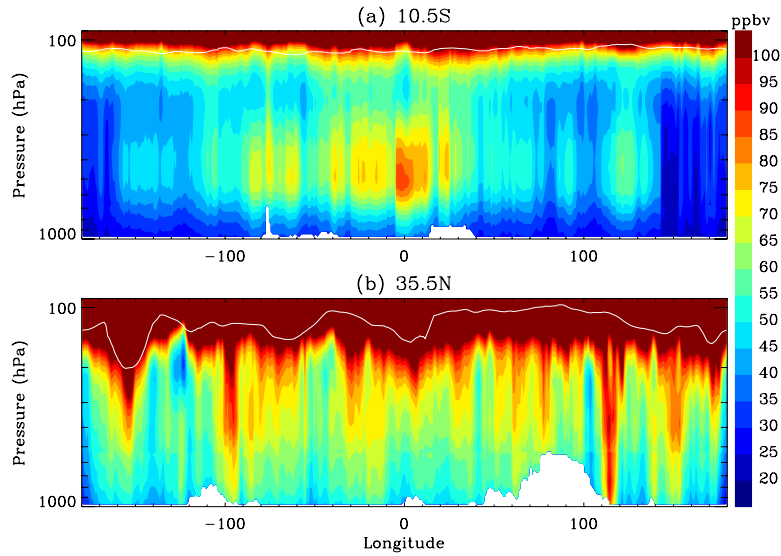


Fig. 9. Longitudinal cross sections of ozone volume mixing ratio from the surface to 100 hPa at 10.5° S and 35.5° N, corresponding to the dashed white lines in Fig. 8. The retrieved partial ozone columns are interpolated to a finer vertical grid and are converted to volume mixing ratio. The white lines indicate the NCEP tropopause.

22737

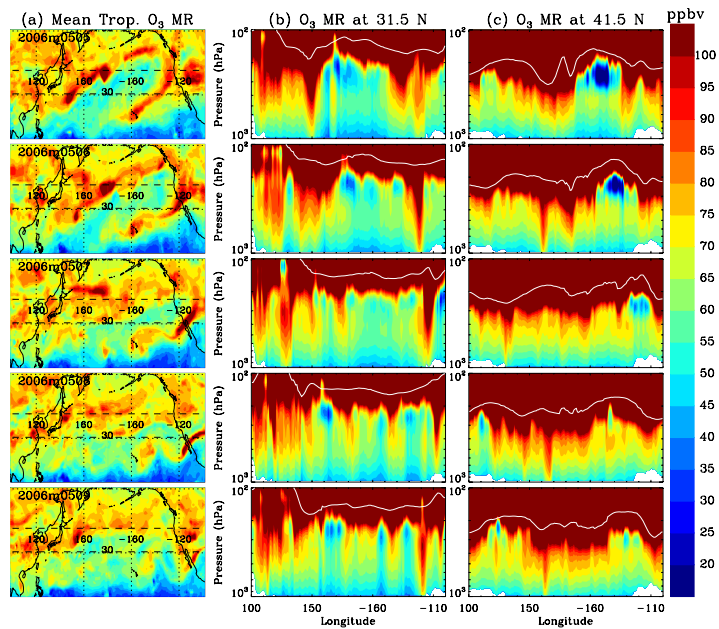


Fig. 10. An event of transpacific transport of pollution from East Asia across the North Pacific Ocean to the United States on 5–9 May 2006. (a) Mean tropospheric ozone volume mixing ratio, (b) longitudinal cross sections of ozone profiles below 100 hPa at 31.5° N and (c) same as (b) but at 41.5° N. The dashed lines on (a) indicate the latitudes of cross sections. The white lines indicate the NCEP tropopause. The data are mapped on 1° longitude×1° latitude grid cells.

22738