Influence of ozone and temperature climatology on the accuracy of satellite total ozone retrieval

L. N. Lamsal,[1,2] M. Weber,[1] G. Labow,[3] and J. P. Burrows[1]

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[1] Deviation of assumed ozone profile shape from true profile in the radiative transfer calculation affects the accuracy of total ozone (TOZ) retrieval. Earlier studies have identified high profile shape sensitivity of retrieved TOZ in polar latitudes, in particular at high solar zenith angles (SZA). This paper is devoted to the question of how TOZ retrievals are influenced by the choice of ozone and temperature profiles from various currently available climatologies. Ozone and temperature profiles from those climatologies are applied in the Weighting Function Differential Optical Absorption Spectroscopy (WFDOAS) algorithm to retrieve TOZ from GOME spectral measurements. Comparison of the retrieved TOZ with ground based measurements from selected stations in the polar, middle-, and low-latitude regions indicate both systematic error and random errors associated with the profile shapes. Those errors become prominent at SZA more than 70°. The systematic error might be caused by the differences in the ozone number density peak altitude between the climatological profile and the actual profile. Biases in true temperature with respect to the climatological temperature profile contribute to the TOZ error by their impact on the ozone absorption coefficient and molecular scattering. Our studies based on GOME spectral measurements and synthetic radiance show that at high SZA, more than 10% systematic error in the retrieved TOZ can be easily introduced by the choice of climatological profiles. The random error is of the same order of magnitude, and it can be related to day-to-day variability of ozone and temperature profiles. In this paper we show that an improved and updated ozone and temperature climatological profiles can reduce the systematic errors in the retrieved TOZ from satellite spectral measurements.

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1. Introduction

[2] Ground and space based instruments have been measuring TOZ for a significant number of years. These measurements do not only provide a unique record of ozone variability on local and global scale but also permit estimation of long-term ozone trends [*Bojkov et al.*, 1995; *World Meteorological Organization*, 1998; *Staehelin et al.*, 2001; *Bodeker et al.*, 2001; *Fioletov et al.*, 2002]. Precise TOZ observations from ground and space are a prerequisite for reliable long-term trend assessments. Ground-based measurements can provide trends at a single site and importantly serve as a control of possible long-term instrumental drifts of satellite instruments. Regular ground-based and satellite measurement comparisons are, thus, needed to ensure that the near global data coverage offered by satellite instruments is of as high quality as possible. Satellite TOZ retrievals often have shown larger disagreement in high latitudes which may complicate trend studies in polar regions.

[3] In addition to the long total ozone record by TOMS and Solar Backscatter UltraViolet (SBUV) starting in 1978 [*Heath and Park*, 1978], the Global Ozone Monitoring Experiment (GOME) [*Burrows et al.*, 1999] has been providing global distribution of ozone for the last 10 years. A long continuous data record length from a single instrument is desirable for long-term trend studies; however, because of the limited lifetime of satellites, a stable and consistent data record has to be derived from multiple instruments [*Bodeker et al.*, 2001].

[4] GOME data is routinely retrieved with the off-line GOME Data Processor (GDP), which has undergone several years of progressive refinements since its first release in 1995. Various validation activities [*Lambert et al.*, 1999; *GDP V3 VALREPORT*, 2002] helped identify many limitations of the earlier versions of GDP and reduced the large discrepancies with a 2–5% bias at SZA < 70° and 10% at SZA > 70° for GDP2.7 to better than 1% in the current version GDPV4.0 [*Roozendael et al.*, 2006] for most part of

[1]Institute of Environmental Physics and Remote Sensing, University of Bremen, Bremen, Germany.
[2]Now at Department of Physics and Atmospheric Science, Dalhousie University, Halifax, Nova Scotia, Canada.
[3]NASA Goddard Space Flight Center, Greenbelt, Maryland, USA.

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globe except in polar regions [*Balis et al.*, 2006]. GDPV4.0 is a standard Differential Optical Absorption Spectroscopy (DOAS) retrieval where slant columns retrieved from a spectral fit are converted to vertical columns using air mass factors (AMFs) calculated at a single wavelength. However, ozone absorption in the Huggins band is wavelength-dependent. Wavelength dependence of ozone air mass factors become important, in particular at high SZA. This is taken into account by the WFDOAS retrieval [*Coldewey-Egbers et al.*, 2004, 2005] that uses wavelength dependent weighting functions. In this approach vertical ozone column density is directly determined in the spectral fitting. Near global validation of the WFDOAS results has shown that the WFDOAS ozone retrievals are of comparable quality to the ground-based data [*Weber et al.*, 2005]. At polar latitudes, WFDOAS results are improved with respect to GDPV3.0 but the discrepancy with respect to ground-based data still persists. In order to minimize the impact of the wavelength-dependent AMF one can select a representative wavelength for the AMF in the standard DOAS approach [*Burrows et al.*, 1999; *GDP V3 VALREPORT*, 2002].

[5] GOME WFDOAS ozone retrieval error studies [*Coldewey-Egbers et al.*, 2005] have identified the assumed ozone and temperature shapes as an important source of error which could lead to errors of up to 5% in the retrieved TOZ at high SZA. Past GDP validation and delta-validation teams have also pointed out the ozone profile shape, which is used in the AMF calculation, as a cause of ozone retrieval errors. Following recommendations of the validation team, the TOMS V7 ozone profile climatology [*Wellemeyer et al.*, 1997] was implemented in GDPV3 to compute off-line AMFs. The current version GDPV4.0 uses the TOMS V8 ozone profile climatology [*Roozendael et al.*, 2006]. The most recent version of TOMS TOZ retrieval algorithm (TOMS version 8) [*Bhartia*, 2003] has corrected several errors that were discovered in its predecessor version 7 [*McPeters et al.*, 1998]. One of the major upgrades to version 8 is the implementation of the improved TOMS V8 ozone climatology.

[6] The choice of climatological ozone profile shapes for radiance calculations as in the case of WFDOAS or TOMS retrievals or in the compution of AMF as in the case of GDPV4.0 is important. The accuracy of satellite TOZ retrievals rely on our ability to model the propagation of radiation in the atmosphere and the resultant energy measured by the satellite instruments. The back-scattered radiance at a wavelength as measured by the instrument depends upon the actual atmospheric state (e.g., the entire ozone profile shape) from top of the atmosphere to surface. Deviation of assumed atmospheric properties from the real atmosphere (e.g., the standard ozone profiles) considered in radiative transfer calculations results in random errors in derived TOZ. *Caudill et al.* [1997] pointed out that the incorrect simulated radiances at high SZA can result in TOZ differences of up to 6%. *Wellemeyer et al.* [1997] showed that day-to-day variability in profile shapes gives rise to a standard deviation of 10% in TOMS TOZ retrieval. Ozone retrieval errors associated with the assumed profile shape do not only affect the ozone retrieval accuracy but also propagate into subsequent data products which are derived from TOZ.

[7] It is clear that a proper choice of ozone profile climatology is important, especially at high latitudes where profile shape sensitivity of simulated radiance (and weighting functions) and/or AMF is high. One of the important aspects for improving TOZ retrievals from satellite measurements is the proper use of representative ozone and temperature profiles. In this paper, we will analyze TOZ retrieved by using some of the more recent climatologies that are commonly used in the TOZ retrievals. Those climatologies are introduced in section 2. The impact of the climatologies on TOZ retrievals at some selected stations of various climate zones are discussed in section 3. A detailed study has been carried out at one of the high-latitude stations (i.e., Syowa) where the choice of climatology is more important and is presented in section 4. The conclusions are given in section 5.

2. Climatologies Under Study

2.1. IUP Climatology

[8] IUP climatology [*Lamsal et al.*, 2004] provides a total ozone column-dependent climatology of ozone and temperature profiles in 1 km steps up to 60 km. This climatology provides a separate set of profiles for both winter/spring and summer/fall seasons in high and middle latitudes of each hemisphere. No seasonal distinction is made for low-latitude profiles. The ozone climatology was based on ozonesondes (middle and high latitudes), Southern Hemisphere Additional Ozonesondes (SHADOZ) [*Thompson et al.*, 2003] (low latitude), Stratospheric Aerosol and Gas Experiment II (SAGE) II [e.g., *McCormick*, 1987; *Chu et al.*, 1989] (low and middle latitudes) and Polar Ozone and Aerosol Measurement III (POAM III) [*Lucke et al.*, 1999](high latitude) ozone profile data set primarily from 1990 to 2000. National Meteorological Center (NMC) and UK Met Office (UKMO) temperature profile data, which are used in SAGE II and POAM III ozone profile retrievals, respectively, and radiosonde (flown together with ozonesonde) data contributed to the associated temperature profile climatology.

2.2. TOMS V7 Climatology

[9] TOMS V7 profile shape climatology [*Wellemeyer et al.*, 1997] was derived by applying a Principal Component Analysis using balloon measurements and data from SAGE II. The climatology comprises 26 standard profiles of ozone and temperatures: ten for high and midlatitudes and six for the low latitude. No distinction was made between hemispheres. These profiles span TOZ values between 125 and 575 Dobson unit (DU) in 50 DU bins. For each ozone profile, a climatological temperature profile is supplied which was derived from SAGE II coincident NMC data. TOMS V7 standard profiles are expressed in Umkehr layers.

2.3. GSFC Climatology

[10] An updated monthly zonal mean ozone profile climatology has been prepared by NASA Goddard Space and Flight Center [*McPeters et al.*, 2006]. This climatology consists of monthly mean zonal mean ozone values for 18 bands (90°S–90°N), each 10° wide and at altitude intervals of 1 km extending up to 60 km. It was compiled from

ozonesonde data from 0 to 24 km, SAGE or Microwave Limb Sounder (MLS) from 29 km to 60 km and a weighted average in between. This climatology was based on ozone observations from 1988 to 2001. The latest version called LLM (Labow, Logan, McPeters) climatology with an improved merging of the sonde and satellite data is available at (ftp@toms.gsfc.nasa.gov/pub/LLM_climatology).

2.4. TOMS V8 Climatology

[11] This is a recent update of the TOMS V7 ozone profile climatology. It provides total ozone classified ozone profiles for each 10° wide latitude bands and for each month. It is an extension of the NASA Goddard Space Flight Center (GSFC) climatology. This climatology, however, does not provide corresponding temperature profiles like TOMS V7.

2.5. KNMI Climatology

[12] The monthly mean ozone climatology developed at the Royal Netherlands Meteorological Institute (KNMI) consists of zonal mean ozone values and standard deviations for 17 zonal bands extending from 85°S to 85°N each of 10° wide and at 19 pressure levels (1000–0.3 hPa) [*Fortuin and Kelder*, 1998]. This climatology was prepared from ozonesonde of 30 ozonesonde stations from 1000 to 30 hPa and SBUV satellite measurements from 30 to 0.3 hPa. Data from 1980 to 1990 were included in this climatology.

3. GOME Total Ozone Retrieval

3.1. Forward Model: SCIATRAN 2.0

[13] SCIATRAN is a 1-D radiative transfer code designed to allow fast and accurate simulation of radiance as measured by the space-based, airborne, and ground-based instruments. SCIATRAN has become a widely applicable and valuable tool for the retrieval of atmospheric constituents from remote radiance measurements. A new generation of SCIATRAN (version 2.0) has been recently released [*Rozanov et al.*, 2005b].

[14] The SCIATRAN radiative transfer model (RTM) [*Rozanov et al.*, 2002] is the successor to GOMETRAN [*Rozanov et al.*, 1997] that was developed to simulate backscattered intensities and weighting functions to retrieve atmospheric parameters from GOME measurements. It covers the spectral range 240–2385 nm comprising the 8 spectral channels of SCIAMACHY [*Burrows et al.*, 1999; *Bovensmann et al.*, 1999] and uses a spherical approximation to arbitrary order that is important for UV limb geometry [*Rozanov et al.*, 2005a]. For nadir application pseudospherical approximation suffices. Rotational Raman scattering by air molecules has been included [*Vountas et al.*, 1998]. In particular for ozone retrievals, the Raman scattering also depends on the ozone profile shape and must be accounted for [*Coldewey-Egbers et al.*, 2005]. The Ring effect is significant for polarization sensitive spectrometers like GOME and SCIAMACHY.

[15] The different ozone and temperature climatologies have been included in SCIATRAN 2.0. In the radiative transfer calculation, ozone and temperature profiles have to be specified. The new upgrade of the RTM facilitates the choice of following sets of ozone and temperature profile climatologies: (1) IUP-Bremen climatology (ozone and temperature), (2) TOMS V7 (ozone and temperature), (3) TOMS V8 (ozone) and COSPAR International Reference Atmosphere 1986 (CIRA) zonal monthly mean (temperature) [*Fleming et al.*, 1988], (4) GSFC zonal monthly mean (ozone) and CIRA (temperature), (5) KNMI zonal monthly mean (ozone) and CIRA (temperature), and (6) Max Planck Institute (MPI) model based ozone and temperature profiles.

[16] The MPI trace gas climatology was derived from 2D chemical transport model calculations [*Crutzen and Brühl*, 1993] and was originally used for all trace gas retrievals from GOME [*Burrows et al.*, 1999]. The MPI model based profiles are available as zonal monthly means but in the analyses presented here only the mean ozone and temperature profiles of the 20–30°N band for June have been used. The motivation for using MPI profile was to elucidate the error in the retrieved TOZ caused by the application of a single arbitrary profile. Selection of proper ozone and temperature profiles from GSFC, KNMI and CIRA climatologies requires as input information on latitude and month. If the total ozone amount is specified, the selected ozone profile is scaled. A priori TOZ information is required for selecting TOZ based climatologies (IUP, TOMSV7, TOMSV8). Using this information the ozone profile from IUP, TOMS V7, and TOMS V8 climatologies is generated as follows:

$$L(T) = \frac{\left(T - T^{(1)}\right)}{\left(T^{(2)} - T^{(1)}\right)} \times L^{(2)} + \frac{\left(T^{(2)} - T\right)}{\left(T^{(2)} - T^{(1)}\right)} \times L^{(1)}$$

where adjacent climatological profiles with total column $T^{(1)}$ and $T^{(2)}$ corresponding to the profiles $L^{(1)}$ and $L^{(2)}$, respectively, that brackets the ozone profile L with TOZ amount of T. In case of GSFC and KNMI climatologies, the monthly mean profile can optionally be scaled to the station TOZ amount.

[17] In IUP climatology, profiles are further classified according to season. TOMS V8 climatology provides such profiles for each 10° wide latitude bands whereas in the IUP and TOMS V7 climatologies, the bands are in accordance with climate zones, for example tropics, midlatitude and polar region. As in many other studies, the climate zones are specified by a fixed latitude. However, the meteorological regimes defined by the subtropical and polar frontal positions are not bounded to specific latitudes [*Hudson et al.*, 2003]. A large variation in ozone profiles along the frontal position and a significant jump in profile characteristics across it can result in a profile shape related error of 1% in the low to middle latitudes and 2% in the middle to high latitudes [*Coldewey-Egbers et al.*, 2005]. Most of the climatologies use a smooth transition between different regions and seasons. The IUP climatology is separated in five zones: two polar regions (>60°), two midlatitude regions (30–60°), and the tropics (<30°) with two seasons winter/spring and summer/fall. Linear interpolation of profile shapes are done in 10° wide bands centered at the boundaries and between seasons, here 15 November to 15 December (fall to winter) and 15 May to 15 June (spring to summer). TOMS V7 climatology is treated in a similar way. This climatology does not make seasonal distinctions in the profile shapes.

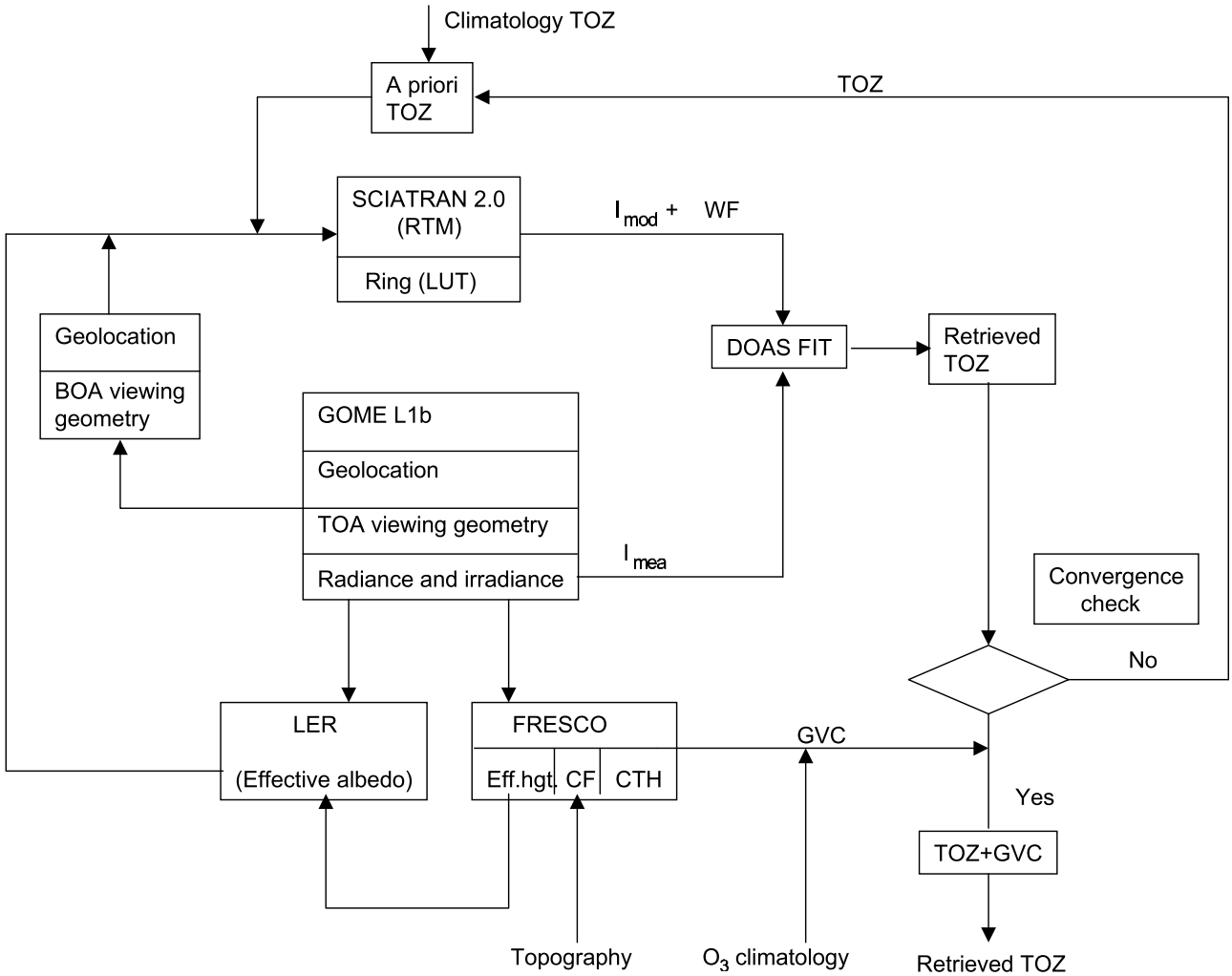

Figure 1. Scheme of the iterative online WFDOAS TOZ retrieval. I_{mod} is the modeled intensity, and I_{mea} is the measured intensity. RTM, radiative transfer model; BOA, bottom of atmosphere; TOA, top of atmosphere; WF, weighting function; LUT, look-up table; LER, Lambert equivalent reflectivity; Eff.hgt, effective height; CF, cloud fraction; CTH, cloud top height; GVC, ghost vertical column; and TOZ, total ozone. See the text for detail.

3.2. Retrieval: Online WFDOAS

[18] Weighting functions are the derivative of the radiation field with respect to the atmospheric parameters and they are used in the retrieval of absorbing species [*Rozanov et al.*, 1998]. *Buchwitz et al.* [2000] introduced the WFDOAS for trace gas retrievals in the near-infrared spectral region of SCIAMACHY. WFDOAS has been applied for the first time to TOZ retrievals from GOME UV spectral measurements [*Coldewey-Egbers et al.*, 2004, 2005] and validation using a large number of ground based measurements indicated an excellent agreement of WFDOAS ozone results with ground based measurements to within 1% for most part of the globe [*Weber et al.*, 2005].

[19] The principle behind this retrieval method is that the measured atmospheric optical depth can be approximated by a Taylor expansion around a reference intensity. A low-order polynomial is added to account for all broadband contributions from surface albedo and aerosol. The Ring effect and the undersampling spectra are treated as an effective absorber. For interfering gases NO_2 and BrO, slant column fitting is applied. This algorithm makes use of wavelength-dependent trace gas weighting functions. This overcomes one of the important limitations of the standard DOAS approach of computing air mass factor (AMF) for the conversion of slant column into vertical column at a single wavelength of 325 nm. The standard DOAS approach assumes that the absorber is weak and the atmosphere is optically thin. In the original version 1 of WFDOAS, the RTM quantities were precalculated and are read from look-up tables (LUT) [*Coldewey-Egbers et al.*, 2005]. The online version of WFDOAS integrates the SCIATRAN RTM into the inversion scheme to retrieve TOZ.

[20] Figure 1 shows a schematic of the online algorithm. The retrieval scheme includes the extraction of GOME level 1b data including geolocation information and viewing geometry, and the preparation of additional data like effective albedo, ghost vertical column, etc., which are required by the retrieval procedure. Cloud top height, cloud fraction, and effective scene height are obtained from Fast Retrieval Scheme for Clouds from the Oxygen A-band (FRESCO)

[*Koelemeijer et al.*, 2001]. Effective height is the weighted sum of ground altitude and cloud top height by the fractional cloud cover. Effective albedo is obtained from GOME Sun-normalized radiance at 377.6 nm.

[21] The algorithm uses nonlinear least squares fitting that includes wavelength shifts and squeeze [*Coldewey-Egbers et al.*, 2005] for the nadir earthshine spectrum to make the direct comparison between measured and modeled backscattered radiances. The modeled radiance, ozone and temperature weighting functions are computed as a function of solar zenith angle, line of sight, relative azimuth angle, surface height and effective albedo by using the radiative transfer model SCIATRAN 2.0 in the pseudospherical approximation.

[22] A 8.2 nm wide fit window from 326.6 to 335.0 nm was selected. Iteration begins with the online simulation of radiance and weighting functions leading to the direct fitting of vertical ozone column. The correct profile shape is chosen from climatology on the basis of the geographical information, day of year, and optionally the TOZ first guess. In order to account for the Ring effect a proper Ring data suited to the atmospheric scenario is selected from a LUT. The LUT was prepared for various atmospheric scenarios and viewing geometries. The impact of cloud in the retrieved TOZ is accounted for by adding a so-called ghost vertical column (GVC) derived from zonal monthly mean ozone climatology, here the GSFC climatology. GVC is the amount of hidden ozone below cloud top pressure assuming an optically thick cloud and is weighted by cloud fraction before adding to the retrieved column. Zonal monthly mean climatologies generally are better suited for tropospheric ozone.

[23] The various climatological ozone and temperature profiles were used in the GOME retrieval. Retrievals were performed at 6 stations: two each from Arctic and Antarctic and one each from middle- and low-latitude regions. Those stations are Resolute (75.2°N, 74.7°E), Sodankylä (67.4°N, 26.6°E), Halley Bay (73.5°S, 26.7°W), Syowa (69.0°S, 39.6°E), Hohenpeissenberg (47.8°N, 11.0°E), and Singapore (1.3°N, 103.9°E). For this comparison, only those GOME pixels were considered whose footprint center lay within 300 km collocation radius from the station, except for Hohenpeissenberg where the collocation radius was reduced to 160 km. This change was intended only for allowing more data in the comparison for other stations. Since the GOME footprint for most part is 320 km across track, the dependence on the collocation radius is not a critical issue. For all stations, the measurement had to take place on the same day of the satellite overpass.

[24] The comparison at Resolute, Sodankylä, and Hohenpeissenberg was done with Brewer measurements. A properly calibrated Brewer instrument provides TOZ values at an accuracy of ±2.5% and its precision is estimated to be ±0.25% [*Kerr and McElroy*, 1995]. The TOZ amount derived from the standard algorithm results in some systematic errors, for instance, coming from uncertainties in the cross sections [*Kerr*, 2002]. Syowa, Halley Bay, and Singapore TOZ observations were from Dobson instruments. The Dobson instrument has performance similar to a Brewer spectrophotometer. Brewer measurements have the advantage over Dobson that they are less sensitive to ozone temperature variation [*Kerr*, 2002; *Weber et al.*, 2005; *Bernhard et al.*, 2005]. In the WMO standard retrieval the ozone cross section is fixed at −46°C that can lead to systematic errors in the Dobson retrieval [*Bernhard et al.*, 2005].

[25] Figure 2 (left) shows the difference between GOME WFDOAS and Hohenpeissenberg daily averaged Brewer TOZ data (in percent) plotted as a function of day in the year 1996–2003. During the period Brewer TOZ ranged from 219 DU to 466 DU and the GOME SZA was in the range 25–73°. Except for the MPI profile for June and 20–30°N latitude band, WFDOAS results show mean biases of less than 0.5% and a small seasonal signature irrespective of which climatology was used. Tropical ozone profiles in June might differ significantly from the profiles at Hohenpeissenberg (midlatitude) and consequently resulted in somewhat larger biases. These comparisons have indicated that the ozone and temperature profiles that are used in forward models to compute the radiance and weighting functions have a minor error contribution to TOZ retrieval at midlatitudes. Similar results are observed in Singapore (Figure 2, right), a low-latitude station. The comparison for this station is limited to the period 1996–2000 because of the problem in the station data quality (V. Fioletov, Meteorological Service of Canada, personal communication, 2005). GOME total ozone retrieved using the MPI profile is slightly lower than those values retrieved using the other climatologies. Note that the effect at Singapore is opposite to that at Hohenpeissenberg. The MPI profile peaks at higher altitude than the actual ozone profiles at Hohenpeissenberg. Its use in the retrieval results in higher TOZ. The MPI profile peaks at lower altitude than the actual ozone profiles at Singapore and its use causes lower TOZ.

[26] Unlike at Hohenpeissenberg and Singapore, WFDOAS results obtained by using different climatologies at high-latitude stations are highly sensitive to the choice of climatological profiles. The difference (in percent) between WFDOAS and the ground-based data from Syowa as a function of day of year (1996–2003) is shown in Figure 3. Each panel corresponds to a retrieval using the indicated climatology. The third dimension is introduced through colors to identify the GOME TOZ dependence by GOME SZA and Dobson TOZ. Each colored circle represents a collocated single measurement and the solid black line the monthly mean of the difference. In Figure 3, the austral polar night as indicated by the GOME data gap separates two distinct seasonal features: (1) summer/early fall with low scatter pattern and (2) late winter/spring with high scatter pattern. Large standard deviation in the spring months is due to the large day to day variability in the atmospheric profile shapes and hence results in larger error in the satellite retrieval. This error is termed as random error and can be as large as 10%. We also identified some systematic errors which are specific to a given climatology application in the respective TOZ retrievals. As can be seen in Figure 3, this error is SZA-dependent which is positive for TOMS V7, negative for MPI and almost zero for IUP climatology. On the basis of these data, a clear dependency on TOZ is not evident. Results from another Antarctic station Halley Bay are fairly similar.

[27] Comparison of retrievals at one of the Arctic stations Sodankylä is shown in Figure 4. Effect of climatology in the retrieval is not noticeable for GOME SZA up to about 75°.

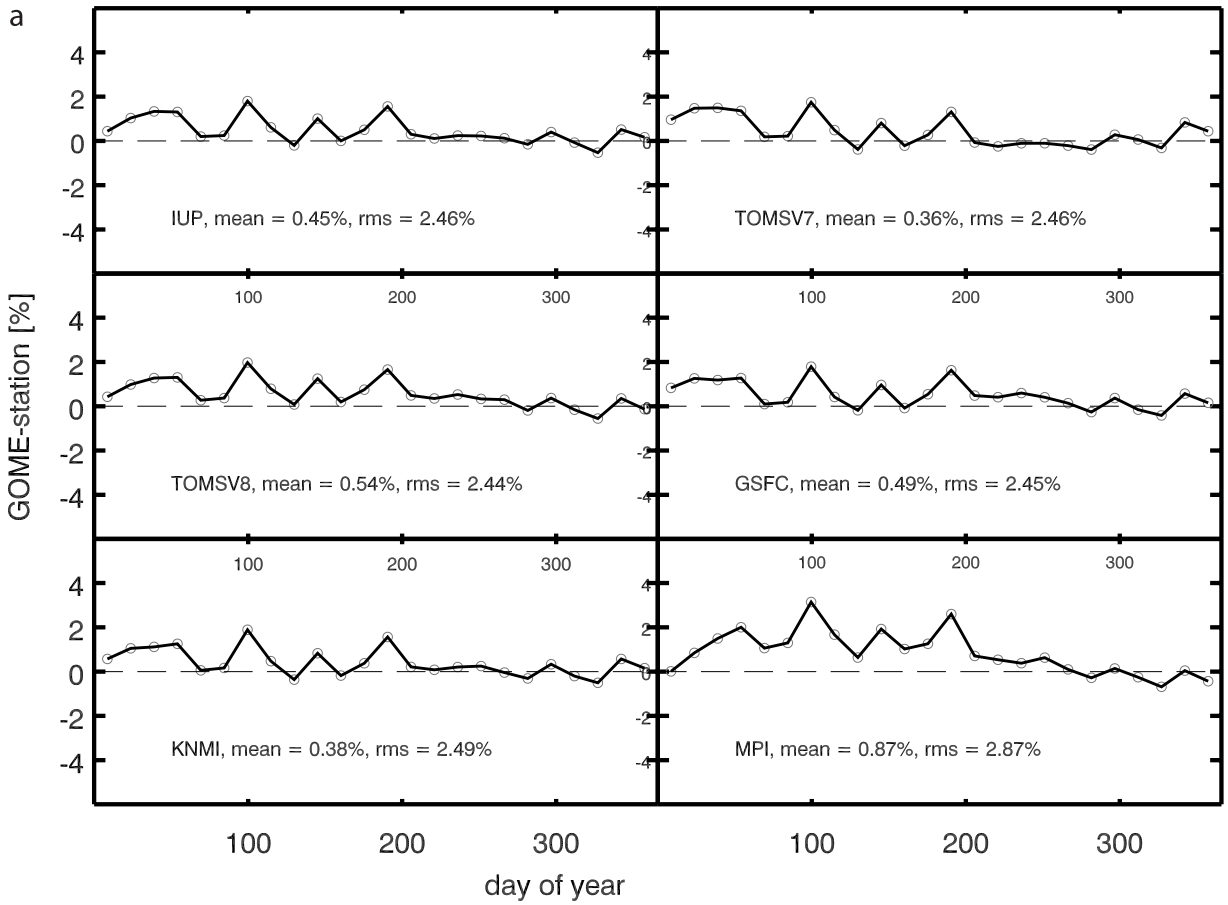

Figure 2. Difference in retrieved TOZ utilizing different ozone and temperature profile climatologies as indicated and Brewer data plotted as a function of day of year (a) at Hohenpeissenberg and (b) at Singapore. The circles are the mean difference averaged over 15 days. The comparison was based on data from 1996 to 2003 for Hohenpeissenberg and from 1996 to 2000 for Singapore.

Differences are observed for higher SZA, in particular for TOMS V7, TOMS V8, and MPI (June, 20–30°N) profile. Large bias of the order of 20% is observed with the MPI profile at high SZA. This result is consistent with that observed at Syowa. Results from another Arctic station Resolute are also similar. Zonal monthly mean climatologies (GSFC and KNMI) result in very good retrievals at both Arctic stations.

[28] In both examples presented above, it is interesting to note that GOME observations at high solar zenith angles are occurring near the polar night terminator (winter) and during polar summer. Near the northern polar nights GOME observations over Sodankylä did not exceed 80° SZA and therefore the SZA-dependent bias is not very prominent for all climatologies. For all zonal monthly mean climatologies including the single MPI profile, the retrieved TOZ at high SZA is negatively biased at both stations (polar night and summer). Near the polar night terminator the TOZ retrieved with the TOMS V7 climatology shows no significant bias at Sodankylä (Northern Hemisphere), but positive bias at Syowa (Southern Hemisphere). A weak negative bias at Sodankylä and no significant bias at Syowa are evident at high SZA during polar summer. These inconsistencies could be due to the difference in ozone and temperature profiles between two hemispheres and/or seasons, which the TOMS V7 climatology does not distinguish.

4. Analysis of Retrievals at Syowa

4.1. Direct Comparison of Sonde Measurements With Climatologies

[29] Four years of ozonesonde data (1996–1999) from Syowa, Antarctica, were used in order to check if the climatologies presented in section 2 can reproduce the measurements. Syowa uses Japanese sonde KC which is similar to the Carbon Iodide (CI) sonde of *Komhyr* [1969]. KC ozonesondes give less consistent results than Electrochemical Concentration Cell (ECC) [*Komhyr*, 1969; *Smit et al.*, 1998], however, this station continues using the sensor for reasons of homogeneity of long-term ozone profile time series. Dobson spectrophotometer stationed at Syowa provide matching total ozone values to ozonesonding profiles. TOZ measurements from Dobson spectrophotometer have the relative uncertainty of 2% [*Basher*, 1982; *Fioletov et al.*, 1999]. Both of these data sets were available from the World Ozone and Ultraviolet Radiation Data Center (WOUDC). In order to compare ozone concentration from ozone column classified climatologies (IUP, TOMS V7, and TOMS V8)

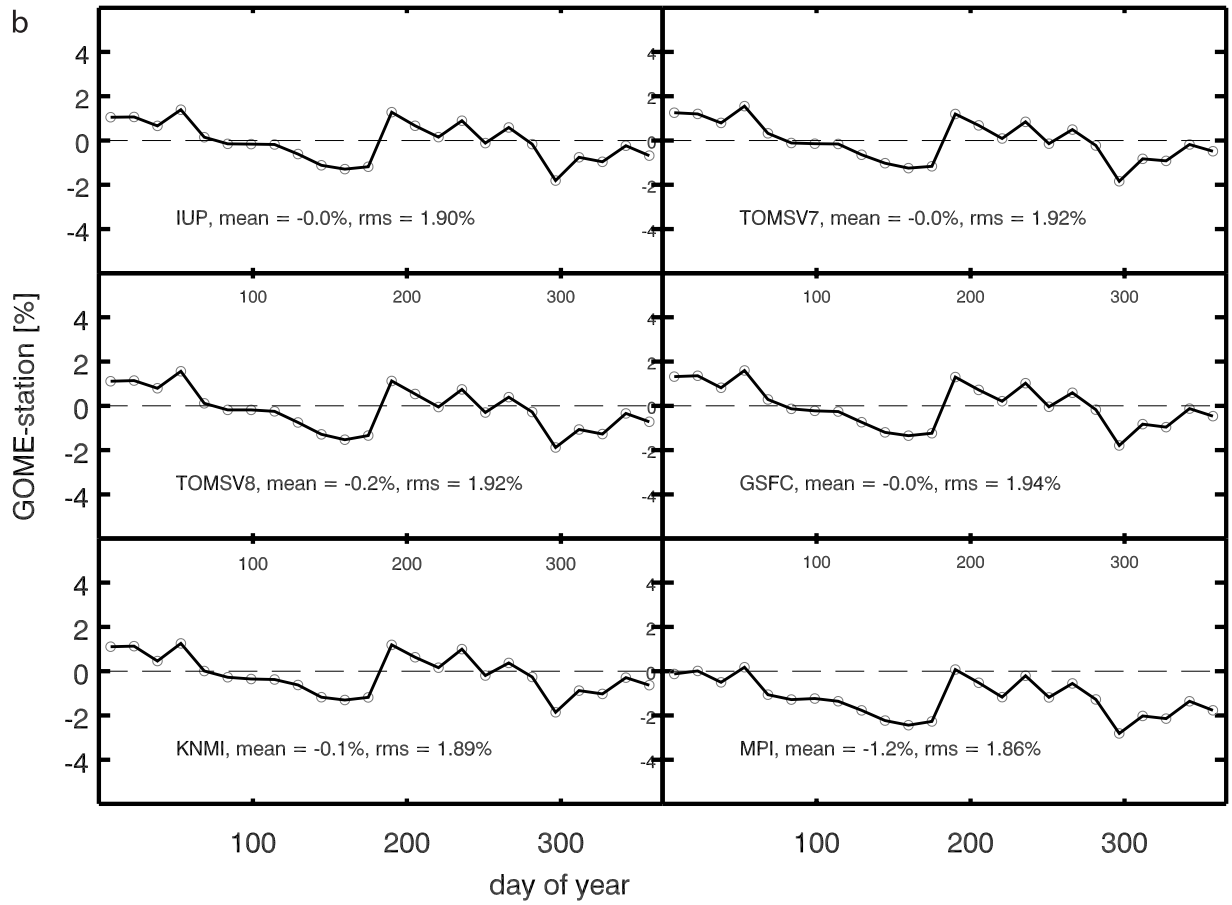

Figure 2. (continued)

with that from ozonesonde measurements, TOZ information obtained from Dobson instrument is required. Linear interpolation in TOZ is essentially performed using the equation given in section 3. TOMS V8 climatology requires additional information on month and latitude. For the zonal monthly mean climatologies (KNMI and GSFC), month and latitude information suffice. Also, scaling of the zonal monthly mean profiles by the station TOZ amount is possible. Given these variables (TOZ, month, latitude), climatological ozone concentration at any altitude level can be compared with regular ozonesonde measurements.

[30] For ozonesonde-climatology comparison, we ensure that ozone concentrations are expressed in the same units and the values are on a common grid. Both IUP and GSFC climatologies provide ozone values at 1 km altitude steps. KNMI climatology expressed in pressure levels was converted to the same altitude levels by using the US standard atmosphere and spline interpolation. Special care was taken to express the TOMS V7 and TOMS V8 ozone values in Dobson units given for Umkehr layers into the same altitude intervals as IUP climatology. Ozone number density at layer midpoints were calculated from the ozone values at consecutive layers which were finally interpolated to the 1 km altitude grid. Between 60 km and 98.6 km the volume mixing ratio at 60 km was linearly interpolated to 0.0521105 ppmv at 98.6 km from *Liang et al.* [1997]. The individual ozonesonde readings in units of partial pressure were converted into number density.

[31] Figure 5 gives the ozone number density reproduced by the climatologies at different altitudes as a function of TOZ amount measured at Syowa. Sonde data interpolated to those altitude levels are also shown in Figure 5. Shown are the mean and 2σ variance of ozone from averaging in 25 DU wide TOZ bins. The selected altitudes cover a broad range from the tropopause to the height of ozone peak. It consists of dynamically highly sensitive region (around tropopause) where short-term ozone variation is controlled to a large extent by horizontal and vertical transport [*Salby and Callaghan*, 1993] as well as altitudes where the chemical destruction of ozone is rapid and severe. Higher variability in ozone is observed in this altitude range.

[32] Climatological ozone number density is less than ozonesonde values at 10.5 km. IUP climatology shows better agreement than others but it also underestimates winter/spring ozone values. All updated and recent climatologies reproduce ozonesonde values at 15 km and 19.5 km very well. KNMI climatology overestimates observations mainly in the lower stratosphere. This climatology is based on data from 80s and the dramatic ozone loss seen mostly in the 90s is not captured. TOMS V7 climatology appears to be too low around and below 15 km. Simple scalar scaling of the monthly mean climatologies slightly improves their agreement with ozonesondes.

[33] The reproducibility of the climatologies for ozone number density presented here only at selected altitude levels should not be understood as the general feature at

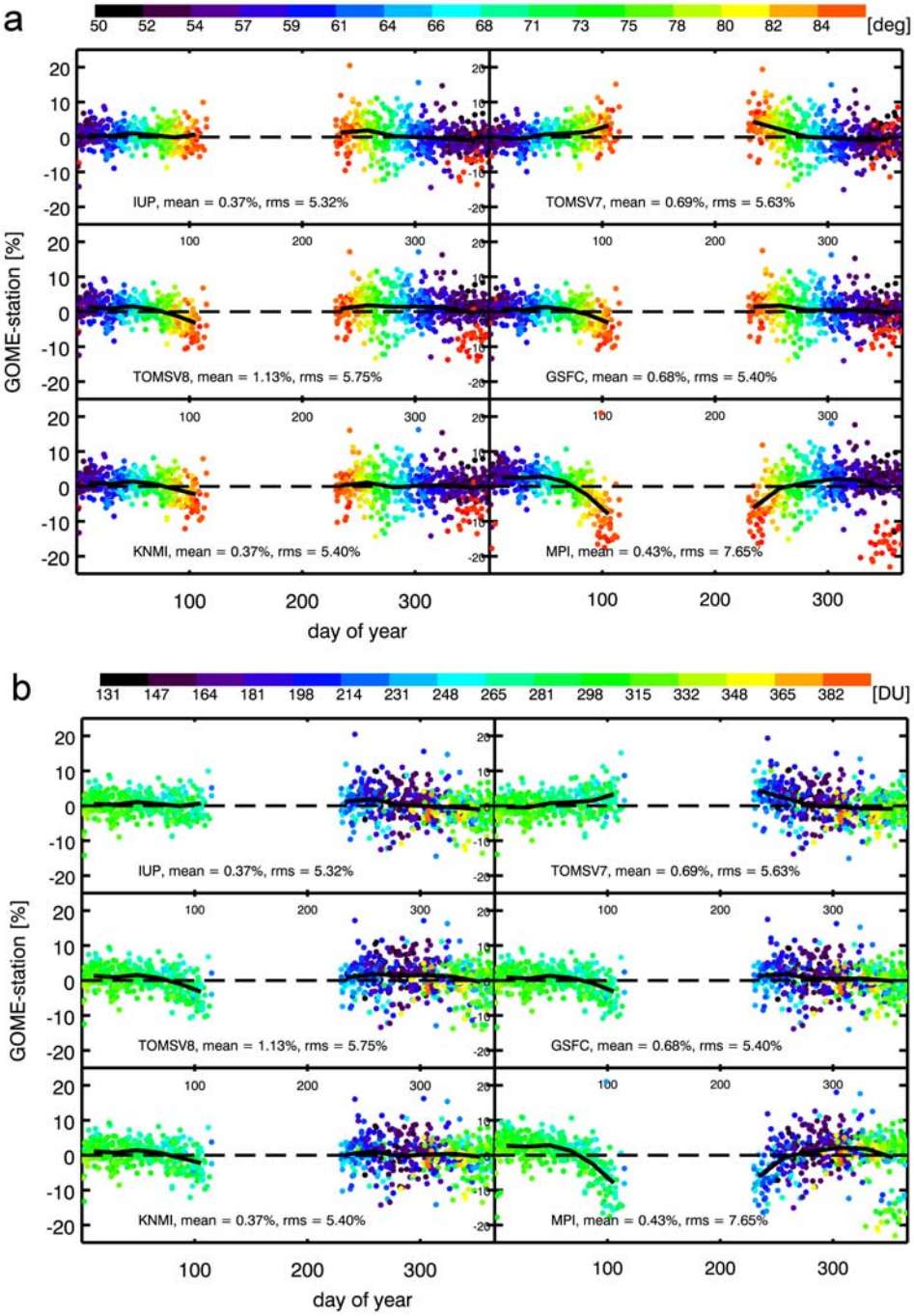

Figure 3. Difference in retrieved TOZ and Dobson data plotted as a function of day of year at Syowa. Ground based TOZ data were obtained from a Dobson spectrometer. The data points are color coded by (a) GOME SZA and (b) Dobson TOZ through the use of colors.

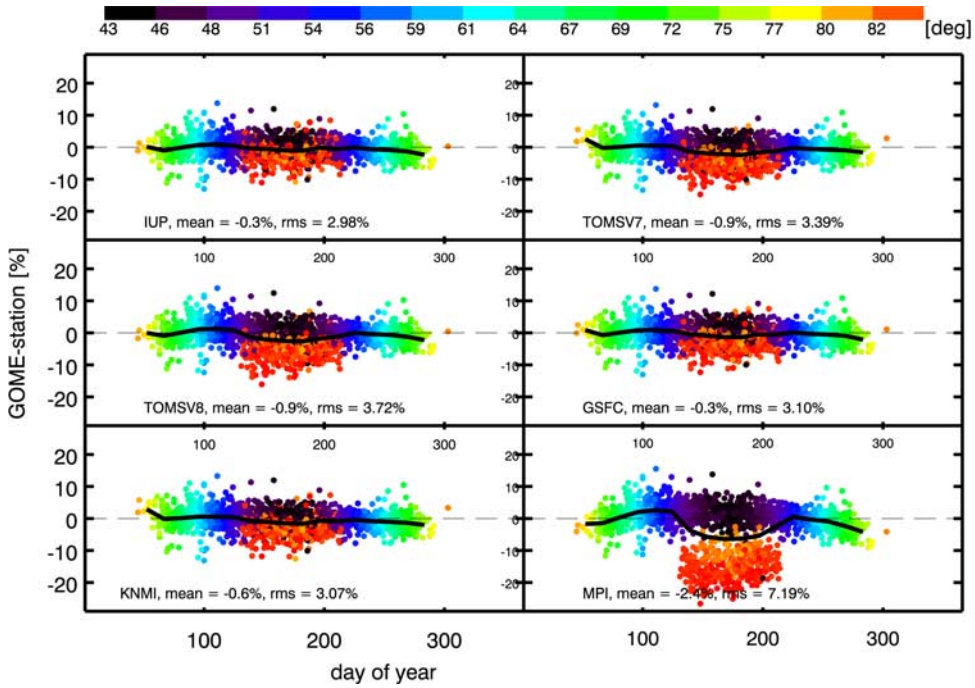

Figure 4. Same as Figure 3 but for Sodankylä.

all levels. A climatology performing well at a certain altitude level may not behave as well at other levels. Additionally, the test was limited to the altitude below 24 km. Although the altitude region of ozone number density peak contributes most strongly to the total column amount, TOZ retrieval accuracy rely on the accuracy of a climatological profile over the entire altitude range. Therefore it is difficult to draw any firm conclusion regarding the accuracy of a particular climatology and its impact on TOZ retrievals. Nevertheless, for the purpose of identifying the profile shape related errors in TOZ retrieval this kind of test might be helpful to some extent. In general the agreement is satisfactory at the ozone number density peak height for all climatologies, with the exception of the KNMI and TOMS V7 climatologies. This could be the reason for such larger errors observed in the case of TOMS V7 climatology during high solar zenith angle conditions (Figure 3). Despite the large discrepancies in ozone number density between ozonesonde and KNMI climatology, the TOZ retrievals with it is hardly any worse. This could primarily be due to the fact that the GOME SZA is moderate when there is extremely low ozone and consequently larger ozonesonde-KNMI difference (see Figure 3). Relatively good ozonesonde-climatology agreement at those selected altitudes for IUP climatology is suggestive of improved retrievals upon its use.

4.2. Retrieval Studies Using Modeled Radiance

[34] For further investigation of the influence of ozone and temperature profiles of various climatologies on TOZ retrievals, we also retrieved total ozone from synthetic radiances. The main motivation here is that all the input quantities which produce the spectra will be known and the role of each of the inputs on the retrieved TOZ can be investigated.

[35] The radiances were computed using SCIATRAN 2.0 at TOZ 100 DU, 150 DU, 200 DU, 250 DU, 300 DU, and 350 DU, SZA 84°, 80°, and 60°, relative azimuth angle 0°, line of sight 0°, surface height 2 km, effective albedo 0.1, and latitude and longitude that of Syowa. A clear sky scenario was assumed. Ozone and temperature profiles were taken from IUP climatology. These radiance were used to retrieve TOZ using the online WFDOAS algorithm. The main purpose of taking the same radiative transfer model is to avoid any retrieval error from model bias. We retrieved TOZ from the synthetic radiances by altering the climatologies but keeping all other input parameters identical. For zonal monthly mean climatologies GSFC and KNMI, two retrievals were performed: (1) with and (2) without scaling ozone profiles to the retrieved TOZ during each iteration. Figure 6 shows the error in retrieved TOZ plotted as a function of total ozone. Results are presented for three SZA 84°, 80°, and 60°. As expected, the retrieval using IUP climatology reproduced TOZ (i.e., almost zero bias). TOZ using TOMS V7 climatology shows systematic bias beyond 200 DU and is SZA dependent. The error can be as high as 10% at 350 DU and SZA of 84°. Retrieved TOZ using TOMS V8 climatology is biased for low ozone cases and the error can reach up to 5% at 100 DU and SZA of 84°. Errors from zonal monthly mean climatologies range from −2% to 2% depending upon the TOZ amount. They tend to underestimate at low- and high-ozone cases. The method of scaling zonal monthly mean ozone profiles to total ozone can improve the retrieved TOZ by up to 1% in those cases. Retrieved TOZ using MPI profiles largely underestimates and the error enhances with SZA.

[36] The observed discrepancies in the TOZ retrieved by using different climatologies have indicated that the distribution of ozone with altitude can be different despite having

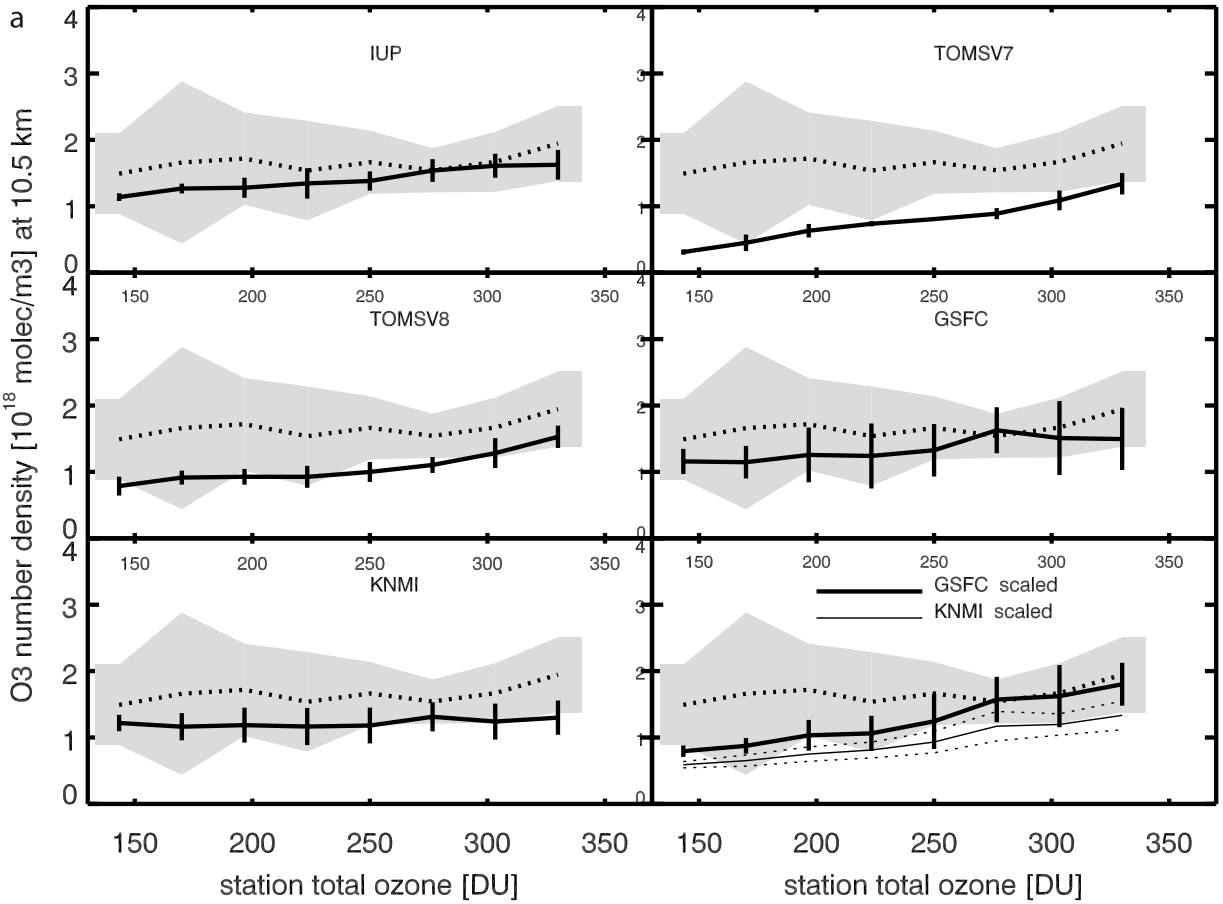

Figure 5. Ozone number density at (a) 10.5 km, (b) 15 km, (c) 19.5 km, and (d) 24 km plotted as a function of ground based TOZ measurements at Syowa (69°S, 39°E). The dotted line represents the mean, and the shaded region indicates 2σ level of ozonesonde measurements. The solid line and the bar represent the mean and 2σ level of ozone values reproduced by the climatologies. In case of GSFC and KNMI climatologies, the profiles scaled to the station TOZ amount are also shown in a separate plot (sixth figure in each panel). Here the 2σ level of ozone values for the KNMI scaled are shown by two thin dotted lines.

the same ozone column amount. The top of atmosphere (TOA) earth shine radiance responds differently to changes in ozone concentration and temperature by ozone absorption at various altitude levels [*Rozanov et al.*, 1998]. As an example, we further analyzed the TOZ retrieval at 250 DU of Figure 6. Figure 7 shows the ozone and temperature profiles in September at 69°S from various climatologies. IUP, TOMS V7 and TOMS V8 ozone profiles correspond to the ozone column amount of 250 DU. Both IUP and TOMS V8 profile peak at around 20 km, TOMS V7 around 17 km, and MPI profile at 25 km. Zonal monthly mean profiles from GSFC and KNMI show ozone depletion near 20 km as might be expected from simple averaging of all profiles in September, which consists of ozone hole profiles as well. Moreover, some ozone profiles are flatter than others. Temperature profiles also vary significantly, particularly the TOMS V7 temperature profile which is warmer than others up to 35 km and colder above. This difference might be due to the fact that TOMS V7 climatology provides a set of temperature profiles common to warmer Northern Hemisphere and colder Southern Hemisphere. The tropical MPI profile is warmer and is characterized by a sharp tropopause [*Randel et al.*, 2003].

[37] Figure 8 shows the ozone and temperature weighting functions at 327.9 nm, which corresponds to the retrieved TOZ as following: IUP 245.2 DU, TOMS V7 254.9 DU, TOMS V8 246.4 DU, GSFC 249.1 DU, KNMI 247.9 DU, and MPI 240.0 DU. Ozone profile corresponding to the 250 DU TOZ (input) spans from 0 km to 60 km. The difference between input 250 DU and TOZ retrieved with the IUP climatology is mainly due to the fact that the surface altitude is assumed to be 2 km for TOZ retrievals. Integrated ozone from 0 km to 2 km turns out to be about 4.7 DU. The TOMS V7 profile thus can cause an overestimation of about 3.9% (254.9 DU vs 245.3 DU), the MPI profile causes underestimation by about 2.2% (240.0 DU vs 245.3 DU) and all other show better agreement.

[38] The ozone weighting function indicates the percent change of the radiance field due to a 1% change in the vertically integrated ozone profile. Likewise the temperature

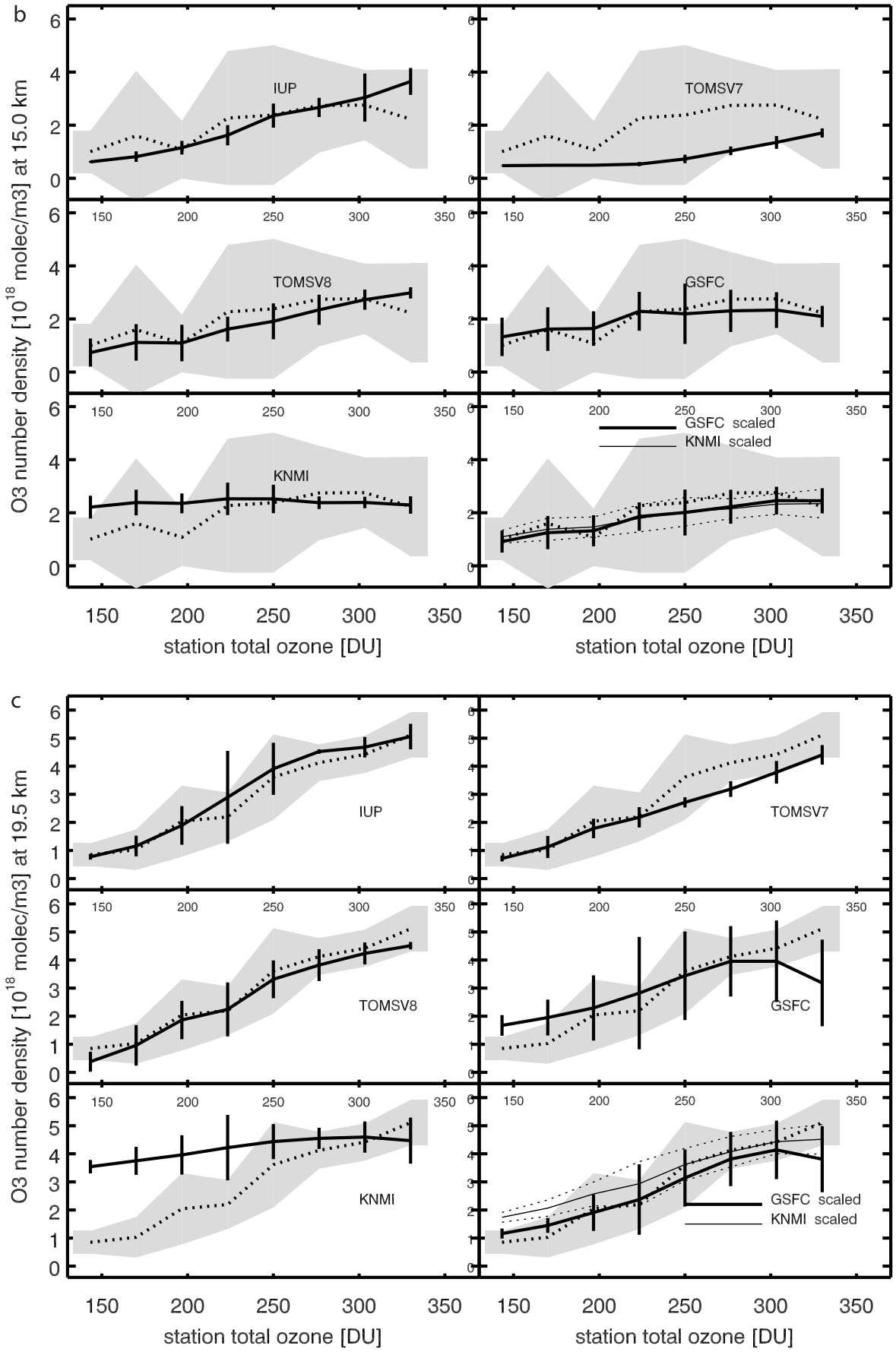

Figure 5. (continued)

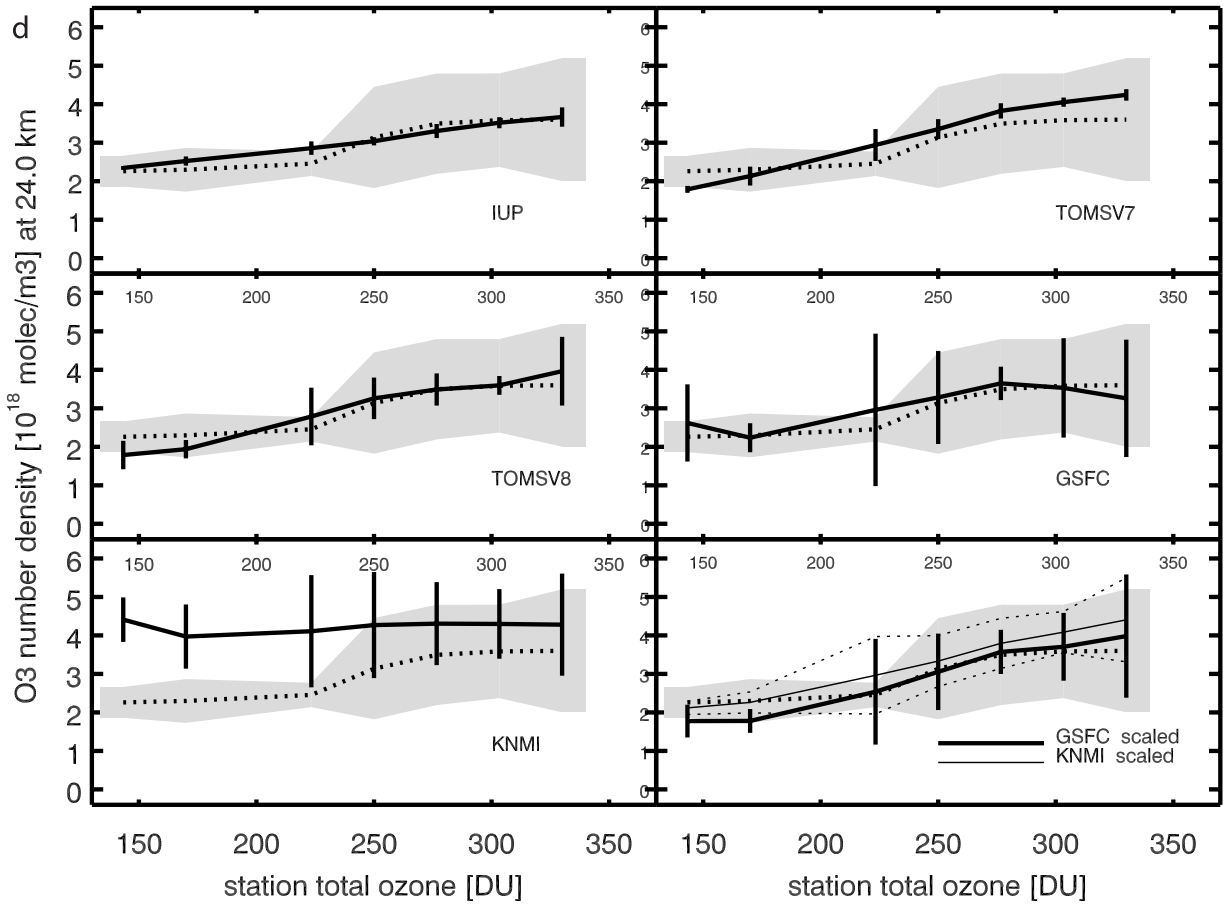

Figure 5. (continued)

weighting function represents the change of radiance field due to a change of 1 K at all altitudes. The shape of these weighting functions might differ significantly depending on the climatology as presented in Figure 7. Ozone weighting functions from various climatologies resemble their respective ozone profile shapes. TOMS V7 TOA radiance is associated with broad high response altitude range peaking at about 17 km. For MPI, the response is mainly coming from 20 to 35 km with maximum response from about 25 km. TOMS V8, GSFC, and KNMI climatologies have a similar peak response altitude around 23 km. These monthly mean climatologies have a stronger secondary maxima that peaks around 37 km. Their lower response at lower altitudes might be compensated at higher altitudes. Despite the fact that the temperature weighting functions from different climatologies also vary from each other, most significantly for TOMS V7 and MPI, the relative shapes are quite similar. Influence of temperature on molecular scattering and ozone absorption coefficient are the cause for the larger values in lower altitude and higher altitude respectively. Here it is important to point out that the weighting functions on which the TOA radiance depends is shown only for 327.9 nm. The WFDOAS TOZ retrieval uses spectral fitting over entire wavelength window from 326.5 nm to 335 nm. It is therefore difficult to completely associate the observed difference in the ozone and temperature weighting functions to the retrieved TOZ.

[39] These investigations have indicated that the larger errors from TOMS V7 profile shape climatology might also be related to its temperature profile. To confirm this, we retrieved total ozone from the same synthetic radiance as explained above using ozone profiles from various climatologies but in combination with a single temperature profile from a zonal monthly mean MPI profile of September and 60–70°S latitude band. This provides an opportunity to isolate TOZ error from temperature and from ozone of various climatologies. No significant difference is found in the case of zonal monthly mean climatologies (GSFC, KNMI, and TOMS V8), some discrepancy is found in the case of IUP climatology, and a substantial difference is evident for TOMS V7 climatology (Figure 9). As discussed in section 3, the CIRA zonal monthly mean temperature climatology has been used in combination with GSFC, KNMI, or TOMS V8 ozone climatology. Similar results for these climatologies would mean that the MPI profile agrees with the CIRA profile. Retrieval using IUP ozone climatology and MPI temperature profiles also shows an increased error around 300 DU and the error is SZA-dependent reaching up to 2% for SZA of 84°. It could be that the IUP SH (Southern Hemisphere) polar winter/spring temperature profiles for certain ozone class have some errors due to the use of limited data sets in the polar regions [*Lamsal et al.*, 2004]. Retrieval errors with TOMS V7 ozone and temperature climatologies as shown in Figure 9 is consistent with the one shown in Figure 3. The TOZ error, which is both SZA-

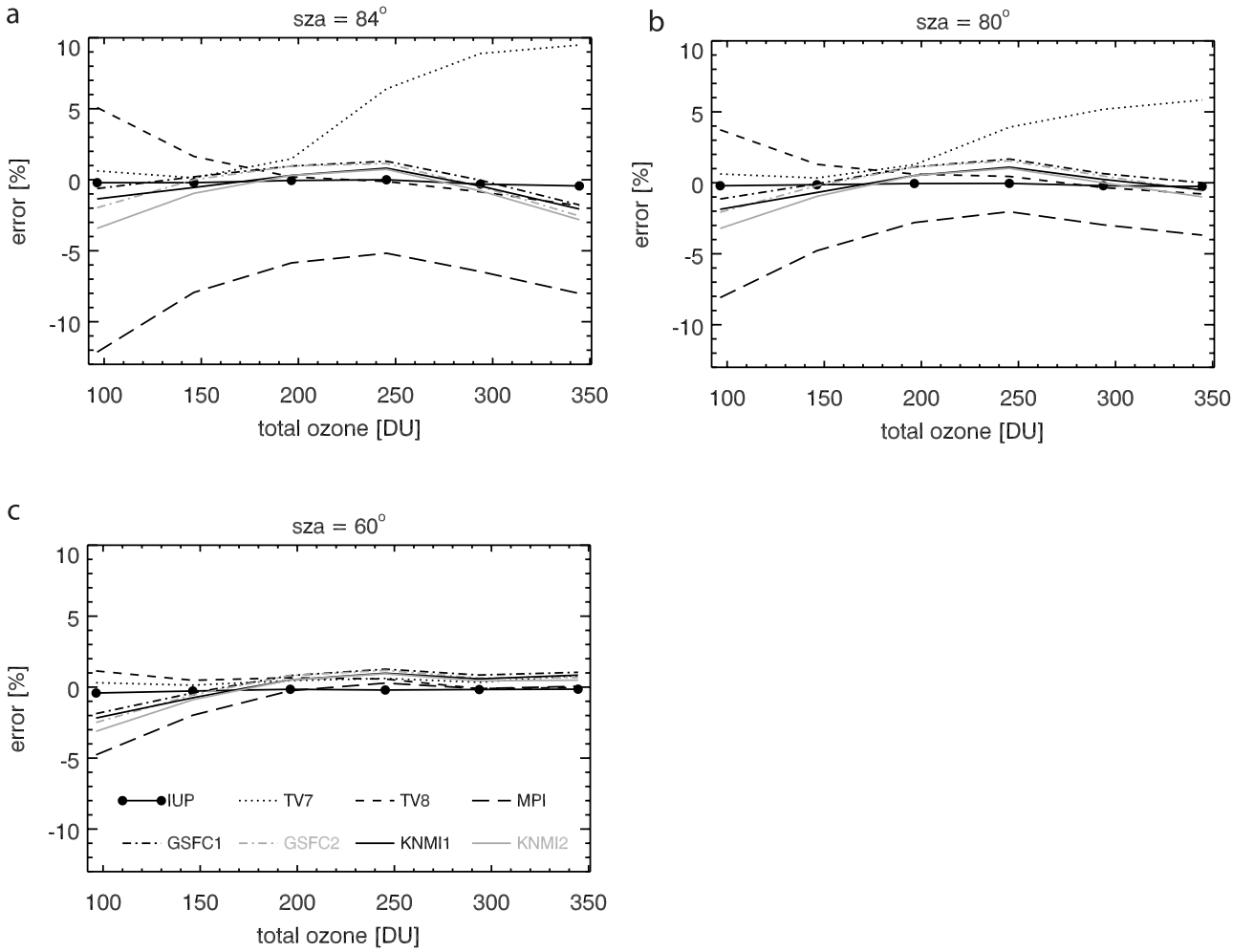

Figure 6. Error in the retrieved TOZ caused by the differences in TOZ amount or/and distribution of ozone and temperature in the above mentioned climatologies using the IUP climatology as a reference. Solar zenith angle of (a) 84°, (b) 80°, and (c) 60°. GSFC2 and KNMI2 represent the original profiles, and GSFC1 and KNMI1 are the profiles which are obtained by scaling the original profiles with a given ozone column.

and TOZ-dependent, can reach up to 10% for SZA of 84° and TOZ of about 350 DU. Interestingly, using TOMS V7 ozone and MPI temperature profile the error in the retrieved TOZ is lowered systematically by almost 50% and as such the TOZ error for SZA of 84° and TOZ of about 350 DU decreases to about 5.5%. Thus it appears that the TOMS V7 temperature is responsible for 50% of the observed error in Figure 3. Note that at lower SZA the error in retrieved TOZ is less than 0.5% irrespective of whether the temperature profile is taken from the TOMS V7 climatology or the MPI model-based climatology.

5. Conclusion

[40] Good a priori knowledge of vertical profiles of ozone and temperature are essential for the calculation of backscattered UV radiances used in the total ozone retrieval. The temperature profile is needed because ozone cross sections are weakly temperature-dependent. Error in assumed profiles propagates into simulated radiances, AMF, etc. thereby finally affecting the accuracy of retrieved TOZ. In order to investigate the profile related TOZ retrieval error, TOZ retrievals were analyzed at 6 stations that represent Antarctic (Syowa and Halley Bay), Arctic (Sodankylä and Resolute), midlatitude (Hohenpeissenberg) and low latitude (Singapore). More stations were included in the polar region because the profile sensitivity of TOZ retrieval is significantly larger in this region. GOME TOZ retrieved by using WFDOAS shows negligible effect of climatological profiles at Singapore and Hohenpeissenberg but considerable systematic differences are observed at high-latitude stations, specially for SZA larger than 70°. Our studies based on the GOME spectral measurements and synthetic radiances that were used for TOZ retrievals using WFDOAS algorithm, have identified both random and systematic errors in the retrieved TOZ originating from the climatological ozone and temperature profiles. The systematic errors can be up to 10% (e.g., TOMS V7 climatology) at high SZA. Profile sensitivity of TOA radiance is strong when the SZA gets large. By using the IUP climatology an improvement in the retrieved TOZ at high SZA was observed. The TOMS V8 climatology shows improvements over its predecessor TOMS V7. Zonal monthly mean ozone climatologies scaled by TOZ result in similar retrievals as the updated column

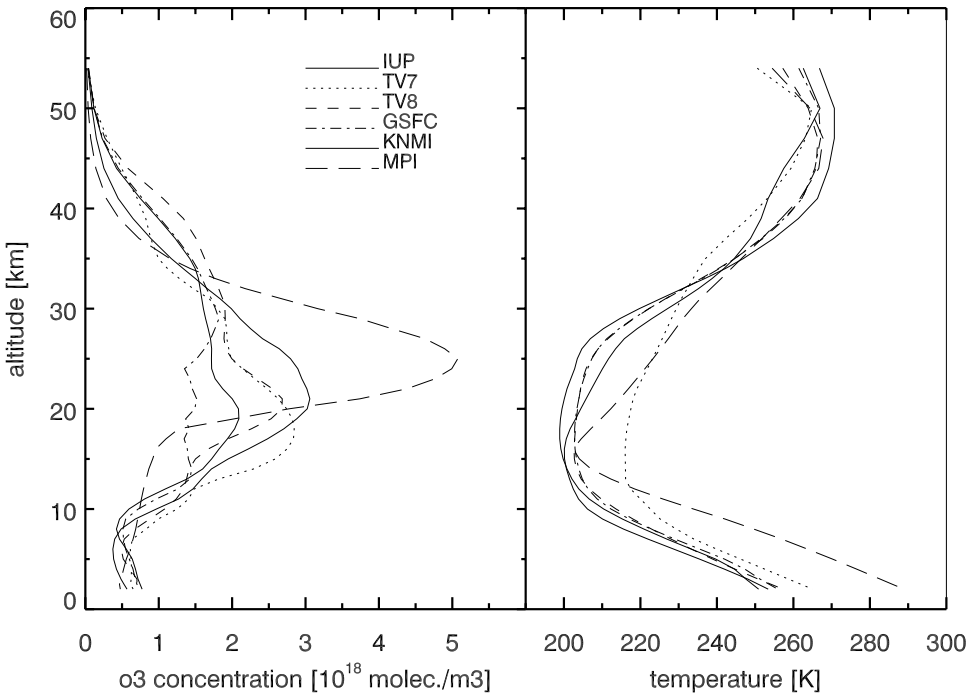

Figure 7. (left) Ozone and (right) temperature profiles in September at Syowa from various climatologies. For TOZ classified climatologies, the profile corresponds to 250 DU.

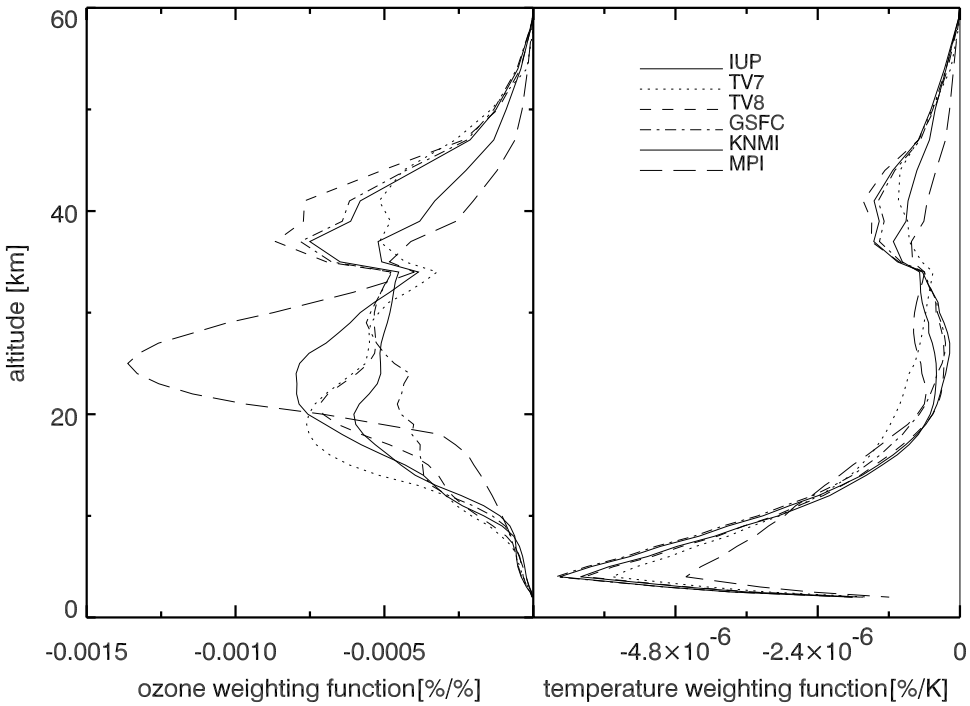

Figure 8. (left) Ozone and (right) temperature weighting functions at 327.9 nm in the Hartley-Huggins band of ozone. These weighting functions correspond to the TOZ retrievals using simulated radiance based on the 250 DU profile of the IUP climatology.

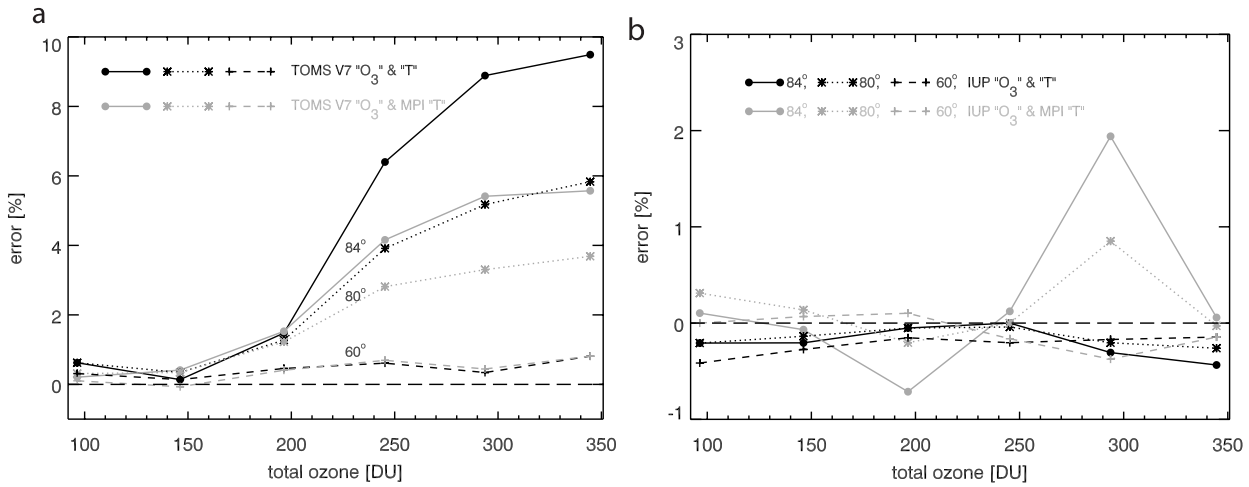

Figure 9. (left) Error in retrieved TOZ associated with TOMS V7 temperature profile. Error in retrieved TOZ using the TOMS V7 ozone and temperature profiles are shown in black lines and the grey lines represent the errors while using TOMS V7 ozone climatology in combination with the MPI temperature profile (of 60–70°S, September). (right) Similar results but for the IUP climatology. Reference scenarios were calculated using the IUP climatology (at the location of Syowa, winter/spring seasons).

classified IUP climatology, with the exception in southern polar region (e.g., Syowa) where the chemical loss of ozone is severe. The investigation carried out in this paper provides a clear message that regular updates of ozone and temperature climatologies can remove some of the discrepancies observed in the current satellite ozone products.

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J. P. Burrows and M. Weber, Institute of Environmental Physics and Remote Sensing, University of Bremen, Otto-Hahn-Allee 1, D-28359 Bremen, Germany.

G. Labow, NASA Goddard Space Flight Center, Greenbelt, MD 20771, USA.

L. N. Lamsal, Department of Physics and Atmospheric Science, Sir James Dunn Building, Room 129, Dalhousie University, Halifax, NS, Canada B3H 3J5. (lamsal@iup.physik.uni-bremen.de)