

Problems regarding the tropospheric O₃ residual method and its interpretation in Fishman et al. (2003)

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In this article we will show that the Total Ozone Residuals (TOR) method as presented by Fishman et al. (2003) contains an erroneous assumption, due to which the TOR as presented in this article reflects mainly a tropospheric O₃ climatology (Logan, 1999), scaled to the layer between the surface and the tropopause height, rather than a satellite measured TOR. We will show that it is possible to obtain a tropospheric O₃ column that is very similar to what is being presented in Fishman et al. (2003), solely based on the Logan (1999) tropospheric O₃ climatology and an estimate for the tropopause heights without using satellite data. Furthermore, we will show that one of the interpretations of observed O₃ variability (high TOR values over Southeast Asia) is not the result of pollution, but primarily of tropopause height variations. We suggest adjusting the method in such a way that the final retrieved TOR product becomes independent of the actual values of the Logan (1999) O₃ climatology. The proposed adjustments are also more in line with the original methodology suggested by Fishman and Balok (1999) in a previous paper. We furthermore show that the assumption that the stratospheric O₃ column is nearly constant within a 5-day period is not always valid. This can introduce errors in the final TOR product, although we recognize that these errors may become small when averaging the TOR product over longer periods of time. However, we feel that a detailed study on this assumption is also needed.

1. Introduction

Over the last decade or so several methods have been developed to estimate the tropospheric O₃ content from satellite observations (for example Hudson and Thompson, 1996; Kim et al., 1996; Ziemke et al., 1998; Fishman and Balok, 1999; Newchurch et al., 2003; Fishman et al., 2003). Observing tropospheric O₃ from space is difficult, because the largest part of the total ozone column is found in the stratosphere, with tropospheric O₃ contributing only a small part to the total O₃ column. Most of

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the O₃ that (backscattered) sunlight has encountered along its path through the atmosphere is located in the stratosphere. As a result, most of the O₃ information in the backscattered sunlight is stratospheric, not tropospheric, and any retrieval from the backscattered sunlight has a reduced sensitivity to the troposphere (Hasekamp and Landgraf, 2002). In addition, clouds often obscure the observation of tropospheric O₃ at ultra-violet and visible wavelengths. One commonly applied method is to use one instrument which measures total O₃ columns and another instrument which measures stratospheric O₃ profiles. Subtracting the stratospheric part from the total O₃ column yields (at least in principle) a residual which is the tropospheric part This is also the methodology followed by Fishman et al. (2003).

In this article we focus on two aspects of the retrieval of Total O₃ Residuals (TOR, also known as Total Tropospheric O₃ Columns or TTOC) as recently presented by Fishman et al. (2003). In Sect. 2 we show that the method applied to derive TOR columns should be interpreted as the monthly Logan (1999) climatology with daily varying tropopause heights, with the differences between the daily TOMS total O₃ column and the once-per-5-days SBUV total O₃ column superimposed on it. In Sect. 3 we show that we can derive similar results as found in Fishman et al. (2003) using the Logan (1999) climatology and the tropopause height variations without making any use of satellite data. In Sect. 4 we also show that the interpretation of what is claimed in Fishman et al. (2003) as pollution observed over northern India and China during northern hemisphere summer is in fact predominantly the tropopause height variation between summer and winter. In Sect. 5 we suggest a slight adjustment that makes the TOR column dependent on the vertical distribution of O₃ in the Logan (1999) climatology rather than on the actual O₃ values of the climatology. The suggested adjustment is also more in line with a previous publication on this topic (Fishman and Balok, 2003). In Sect. 6 we investigate the assumption in the Fishman et al. (2003) method that the stratospheric O₃ column is nearly constant for a given 5-day period. We show that the stratospheric O₃ variability for a 5-day period is too large to be ignored. We end with a summary, discussion and recommendation section (Sect. 7).

2. Calculation of TOR

The method described in Fishman et al. (2003) uses Total Ozone Monitoring Satellite (TOMS, 2003) measurements of the total O₃ column and Solar Backscattered Ultra-Violet instrument (SBUV, 2003) stratospheric O₃ profile measurements (Bhartia et al., 1996). Basically, the method subtracts the stratospheric O₃ column from the total O₃ column. The tropopause height for determining the separation between stratosphere and troposphere is obtained from NCEP 6-hourly gridded (2.5°×2.5°) climate model analysis. One of the problems of this method is that an SBUV vertical O₃ profile measurement for a particular site is available only once every 5 days. TOMS total O₃ columns are available globally on a daily basis. In the Fishman et al. (2003) method it is therefore assumed that the total stratospheric O₃ column amount does not change over a 5 day period. In Sect. 6 we will discuss this assumption further.

The SBUV O₃ profile consists of 12 subcolumns between 0.3–1013 hPa. Subcolumns that could be (partly) tropospheric are layers 1 (1013–253 hPa, 0–10 km), layer 2 (253–127 hPa 10–14.5 km) and layer 3 (127–63.3 hPa 14.5–19 km)). The accuracy of the TOR product depends very much on the error and accuracy of the SBUV profile. According to Bhartia et al. (1996), the SBUV O₃ profile is independent from the a priori information between 1–20 hPa (30–50 km). Outside of this region only the total column amounts (1–0.3 hPa and 20–1013 hPa) can be considered to be relatively free of a priori assumptions. Bhartia et al. (1996) concluded that the SBUV retrieval algorithm placed enhanced amounts of tropospheric ozone into the lower troposphere. Validation showed that only the SBUV total column between the surface and 20–30 hPa were in agreement with non-SBUV observed columns (Bhartia et al., 1996; Ziemke et al., 1998). Validation of the SBUV tropospheric O₃ column layers showed that the tropospheric sub-columns do not agree with observations from radiosonde profiles (Fishman and Balok, 1999). The O₃ column in layer 1 is too low; the O₃ column in layer 3 is too high while the O₃ column in layer 2 is comparable to the observed radiosonde O₃ column. Moreover, Fishman and Balok (1999) claim that the total in-

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egrated monthly mean O₃ column over the 1012–63.3 hPa layer is in agreement with the observed columns. Note that this upper boundary of 63.3 hPa is different from the 20–30 hPa as mentioned in Bhartia et al. (1996) and Ziemke et al. (1998). Fishman et al. (1999, 2003) therefore suggests that the “tropospheric” SBUV O₃ profile should be corrected using the Logan (1999) climatology. The assumption is that, although the Logan (1999) climatology may not provide the correct O₃ concentrations and columns at a given location, the vertical distribution in both space and time are represented reasonably well by the climatology. The climatology can then be used to redistribute the O₃ column values in the lowest three SBUV layers while keeping the total SBUV O₃ amount of the lowest three layers fixed.

The correction as defined and formulated in Fishman et al. (2003) is as follows (see Fig. 6 in Fishman et al. (2003) and according to ACPD interactive comment 3-S386). The retrieval method starts by obtaining the lowest part of twelve SBUV O₃ subcolumns:

Layer 1 : Column A (1000–253 hPa)

Layer 2 : Column B (253–126 hPa)

Layer 3 : Column C (126–63 hPa).

(1)

Next, the Logan (1999) tropospheric O₃ climatology is partitioned into sub-columns that correspond closely to the SBUV sub-layers between 1000 and 100 hPa (the Logan (1999) climatology provides data up to 100 hPa):

Column X (1000–250 hPa)

Column Y (250–125 hPa)

Column Z (125–100 hPa).

(2)

Additional information is needed to extend the Logan (1999) climatology up to 63.3 hPa so that it matches with the lowest three SBUV layers (layers A, B, C in Eq. 1).

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Fishman et al. (2003) assume that the sum of the O₃ in the three layers from the SBUV measurement is correct (in accordance with the findings of Fishman and Balok, 1999). The approach then followed is that the difference between the SBUV columns (A+B+C) and Logan columns (X+Y+Z) is added to the Logan (1999) climatology to represent the “missing” climatology between 100-63 hPa. The “new” column Z* (125–63 hPa) is defined by Fishman et al. (2003) as:

$$Z^* = Z + \Delta Z = Z + (A+B+C) - (X+Y+Z) = (A+B+C) - (X+Y). \quad (3)$$

Next, the ratio of climatological layers (X+Y+Z*) to the sum of the lowest three SBUV layers (A+B+C) is calculated. This ratio is then used to scale the climatological total O₃ between 1000–63 hPa to the observed (SBUV) total O₃ for these layers. The coefficients R₁, R₂, R₃ represent the ratios of the three layers in the extended Logan (1999) climatology and are referred to as the correction ratios of layers 1, 2 and 3.

$$R_1 = X / (X+Y+Z^*) = X / (X+Y+(A+B+C)-(X+Y)) = X / (A+B+C)$$

$$R_2 = Y / (X+Y+Z^*) = Y / (A+B+C) \quad (4)$$

$$R_3 = Z^* / (X+Y+Z^*) = ((A+B+C)-(X+Y)) / (A+B+C).$$

The coefficients R₁, R₂ and R₃ are then multiplied to provide the new layers A*, B* and C*:

$$A^* = R_1 \times (A+B+C) = X$$

$$B^* = R_2 \times (A+B+C) = Y \quad (5)$$

$$C^* = R_3 \times (A+B+C) = (A+B+C) - (X+Y).$$

Note that as a result of putting the entire difference between the SBUV total column (A+B+C) and the Logan (1999) climatology (X+Y+Z) in the missing part of the climatology $\Delta Z = (A+B+C) - (X+Y+Z)$, layers X and Y remain unchanged. Consequently,

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the new layers A* and B* are equal to X and Y, respectively, which are the quantities in the original Logan (1999) climatology. The SBUV measured (A+B+C) sub-columns are thus not effectively redistributed according to the Logan (1999) climatology, which we believe is what the authors aimed for (in accordance with Fishman and Balok, 1999).

5 The new quantities A*, B* and C* are used to calculate the SBUV stratospheric O₃ column (SBUVSOC):

$$\text{SBUV}_{\text{SOC}} = \text{SBUV}_{\text{TOC}} - cC^* - bB^* - A^* \quad (6)$$

in which the coefficients *b* and *c* denote the fractions of layers B* and C* that are located below the tropopause, obtained from NCEP reanalysis data. The tropospheric O₃ residual then becomes:

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$$\text{TOR} = \text{TOMS}_{\text{TOC}} - \text{SBUV}_{\text{SOC}} = \text{TOMS}_{\text{TOC}} - \text{SBUV}_{\text{TOC}} + cC^* + bB^* + A^*. \quad (7)$$

If we now substitute Eq. (5) in Eq. (7) we get:

$$\text{TOR} = \text{TOMS}_{\text{TOC}} - \text{SBUV}_{\text{TOC}} + cC^* + bY + X. \quad (8)$$

Equation (8) has some important consequences. First we assume that the tropopause is located below 125 hPa (which is almost always the case outside of the tropics), so that coefficient *c* becomes zero:

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$$\text{TOR} = \text{TOMS}_{\text{TOC}} - \text{SBUV}_{\text{TOC}} + bY + X. \quad (9a)$$

Equation (9a) consists of two parts. The value *bY*+*X* is nothing more than the Logan (1999) climatology below the altitude at which the tropopause is located. Superimposed on this climatology are the daily variations in total O₃ (TOMS), assuming that TOMS and SBUV measure the same total O₃ columns and because SBUV and TOMS measurements have different sampling times (once per five days/daily). If we assume that the differences in total O₃ columns occur randomly (see Sect. 6), then the differences in total O₃ columns due to the different sampling times would become negligible averaged over a longer period of time (i.e. if the number of samples is large enough).

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As a result, seasonal averages (and probably also monthly averages) primarily reflect changes in the O₃ climatology and tropopause heights.

Alternatively, the tropopause can be located between 125 hPa (15 km) and 63 hPa (19 km). In that case coefficient b becomes one and if we substitute the expression for C^* from Eq. (5) in Eq. (8), we get:

$$\text{TOR} = \text{TOMS}_{\text{TOC}} - \text{SBUV}_{\text{TOC}} + c(A+B+C) + (1-c)(X+Y). \quad (9b)$$

Equation (10b) is a linear combination of the SBUV layers A, B and C, the Logan (1999) climatology below 125 hPa and the daily variations in total O₃ (TOMS). Note that SBUV column C includes a part of the stratosphere. The tropopause height in the tropics rarely exceeds 17 km (90 hPa), so that the value of $(1-c)$ is never small. As a result, also for this case (Eq. 9b) the TOR strongly reflects by the Logan (1999) climatology.

We therefore strongly suspect that, without using any satellite measurements, we should be able to obtain results that in many cases strongly resemble the TOR columns as presented in Fishman et al. (2003), by simply using the Logan (1999) climatology and adding the amount of O₃ between 12 hPa and the tropopause height from a different source (such as a climate model).

3. Total O₃ residual from Logan (1999) and tropopause heights

According to Eqs. (9a) and (9b) the TOR columns in Fishman et al. (2003) are the combination of the Logan (1999) climatology below the tropopause, superimposed with the differences between daily TOMS total O₃ columns and once-per-5-days SBUV total O₃ columns. If differences between SBUV and TOMS total O₃ columns for a 5-day period occur randomly, it is reasonable to assume that these differences are small when averaged over longer periods of time (see Sect. 6). Thus, the TOR product for seasonal averages mainly reflects the Logan (1999) climatology below the tropopause and only marginally reflects tropospheric O₃ as measured by TOMS/SBUV. To further substantiate this point we calculated the TOR using the Logan (1999) climatology and

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the tropopause heights from an ECHAM model simulation on T63 ($\sim 2^\circ \times 2^\circ$ resolution). O₃ concentrations above 100 hPa were assumed equal to the O₃ concentrations in the Logan (1999) climatology at 100 hPa. The 19-layer ECHAM model (Roeckner, 1996) simulation was done using the so-called “nudging” procedure (assimilating ECMWF analysis data) for the year 1996 (Jeuken et al., 1996). The ECHAM model has a vertical coordinate system that follows the surface elevation. The 19-layer ECHAM model has typically a vertical resolution of about 1–2 km in the tropopause region. The tropopause height was defined as the height (vertical gridpoint) for which the vertical temperature gradient was -3° K/km .

Fig. 1 shows the TOR columns from this methodology for the four seasons. The TOR columns show very similar spatial and seasonal variations as the 1979–2000 climatology in Fishman et al. (2003). Differences could be explained by a number of factors: (1) we only used the tropopause height for one year (1996), (2) we used a different tropopause height definition, (3) we did not take into account the fraction of the SBUV column that is added for tropopause heights above 100 hPa as is applied in Fishman et al. (2003), (4) the horizontal resolution of TOMS is about $1^\circ \times 1.25^\circ$ compared to the $2^\circ \times 2^\circ$ resolution from the ECHAM model so that more surface elevation details should be visible in the Fishman et al. (2003) TOR than in our TOR and (5) no systematic differences between daily TOMS and once-per-5-days SBUV total O₃ column measurements are included. However, it is not the scope of this paper to fully reproduce the Fishman et al. (2003) climatology. We merely want to show that the Logan (1999) climatology with some tropopause height estimation results in a TOR column very similar to that in Fishman et al. (2003), as we predicted through inspection of Eqs. (9a) and (9b). Only very little new information about O₃ would be obtained by including the satellite measurements in Fishman’s TOR methodology. Fishman et al. (2003) claim that there is a considerable amount of additional information from the satellite measurements in their TOR, and have substantiated this by comparing the TOR climatology for July with the Logan (1999) climatology total O₃ columns below 250 hPa. However, because the TOR is very dependent on the tropopause height and surface elevation,

comparing the Logan (1999) climatology below 250 hPa (and without taking surface elevation into account) with TOR does not prove that the differences are caused by the satellite measurements. If one takes the surface elevation and tropopause heights into account, which is what we have done in this section, the resulting TOR are very similar to the TOR in Fishman et al. (2003).

4. Interpretation of Asian “summer pollution events”

According to Fishman et al. (2003) and Pierce et al. (2003) the TOR columns show enhanced values during June–August over southern and eastern Asia. Furthermore, enhanced TOR values are seen over the Ganges valley in northern India. It is claimed that this is widespread pollution caused by emissions over northern India. However, when looking in detail at the 1979–2000 TOR climatology in Fishman et al. (2003), we see that this is a persistent feature throughout all seasons. Furthermore, this feature is also present in the “Logan/ECHAM tropopause” TOR in Figs. 1 and 2. There is no such a feature in the actual Logan (1999) O₃ surface concentrations, rather the O₃ concentrations increase along the latitude over India without any longitudinal variations. We therefore conclude that this feature is not caused by pollution, but relates to other processes: a geographical effect and seasonal changes in the TOR values. As noted, the total O₃ column values in the Logan (1999) climatology increase with latitude over southern Asia, but without longitudinal variation. However, there is also a large surface elevation difference between northern India (close to sea level) and the Tibetan plateau (approximately 3–5 km altitude). Therefore, combining the surface elevation changes with the northward increasing O₃ concentrations results in TOR variations that follow local elevation changes. Separate from the influence of the surface elevation however is the question why the TOR column changes so much between the seasons. The O₃ concentrations over southern Asia in the Logan (1999) climatology do not change enough to explain the large differences between summer and winter TOR over India and China. However, large differences between summer and winter TOR are also

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present in the “Logan/ECHAM tropopause” climatology (Figs. 1, 2). The explanation we find by looking at the differences in tropopause height between the winter and summer season (Fig. 3). There is a large change in tropopause height as large as 5 km between winter and summer for this region. This explains the large difference between summer and winter TOR columns over southern Asia: during summer the tropospheric O₃ column is simply about one-third larger than during winter. Moreover, the highest O₃ concentrations are found close to the tropopause, so that a considerable amount of O₃ is added to the tropospheric O₃ column when the tropopause is higher.

We investigate how much O₃ would be needed to explain these TOR differences between summer and winter (differences are 15–25 DU, both in the Fishman et al. (2003) climatology as well as in the “Logan/ECHAM tropopause” TOR). We can convert Dobson Units to mixing ratios as follows:

$$O_3 = F_{DU} \times g \times M_{air} \times (\Delta_{DU}) / (0.1 \times dp \times avo). \quad (10)$$

In which F_{DU} is the conversion factor from molecules/cm² to Dobson Units ($2.69 \cdot 10^{16}$ molecules/cm²), g is the gravity constant (9.81 m/s^2), M_{air} is the molar mass of air (28.64 gr/mol), Δ_{DU} is the difference in O₃ column thickness, dp is the layer thickness in Pa, avo is the Avogadro constant ($6.02 \cdot 10^{23} \text{ mol}^{-1}$) and 0.1 is a factor for the conversion from m² to cm² and kg to gr. If we assume that a 15 DU change occurs in the atmospheric boundary layer, and substitute a layer thickness of 150 hPa (1000–850 hPa which is roughly a thickness of 1.5 km) into Eq. (10), the difference in O₃ concentrations is approximately 125 ppbv (mixing ratio of $1.25 \cdot 10^{-7}$). If a typical O₃ concentration during winter in northern India would be about 50 ppbv (Logan, 1999), then during summer it would be 175 ppbv, which is a very unrealistic O₃ concentration at the surface, even for strong pollution events. On the other hand, a layer of 150 hPa in the tropopause region is similar to a change from 10 to 15 km altitude, which is roughly the tropopause height change from the ECHAM model for this region, and a concentration of 125 ppbv for the tropopause region is very reasonable. The O₃ enhancements during local summer over India and China are not related to pollution, but primarily the

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result of changes in tropopause heights. A summertime maximum in surface O_3 concentrations is not in agreement with our current knowledge, which as also noted by Kim in the ACPD interactive comment 3-S470. Furthermore, this summertime maximum is also not present in the Logan (1999) climatology.

5. Suggestion for improving the TOMS-SBUV TOR column

According to Fishman and Balok (1999), the SBUV total O_3 column amount between 1000 and 63 hPa is approximately correct while the distribution of O_3 amongst the SBUV three lowest layers is not good. In our view the aim of the TOR methodology is to redistribute the 1000–63 hPa column according to the Logan (1999) climatological vertical O_3 distribution. However, the Logan (1999) climatology does not go beyond 100 hPa, whereas the three lowest SBUV layers range from 1000 to 63 hPa. Clearly, some extension of the Logan (1999) climatology is needed to 63 hPa in order to sensibly redistribute the O_3 amongst the three lowest SBUV layers. As indicated in Sect. 2, Fishman et al. (2003) choose to extend the Logan (1999) climatology vertically above 100 hPa by adding the difference between the Logan (1999) climatology total O_3 column and the total of the SBUV three lowest layers to the region above 100 hPa. By doing so, the lower two layers (A* and B*; 1000–125 hPa) are effectively identical to the Logan (1999) climatology values and are not scaled to the SBUV 1000–63 hPa measured column. We believe that this violates the idea of using the Logan (1999) climatology to sensibly redistribute the measured A, B and C columns.

As indicated in Sect. 2, the “problems” arise from the assumption to extend the Logan (1999) climatology vertically above 100 hPa by using SBUV data (Eq. 3). There appears to be no justification for why the O_3 climatology above 100 hPa should be corrected the way it is being done (using the total of the SBUV levels 1–3 minus the Logan (1999) climatology). A more sensible approach could be to extend the Logan (1999) climatology above 100 hPa by assuming the same concentration of O_3 above 100 hPa

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as in the highest Logan (1999) climatology layer. Equation (3) then becomes:

$$Z^* = Z + aZ. \quad (11)$$

With a numerical value for a , which is the thickness of the layer between 100 and 63 hPa relative to the thickness of the layer between 125 and 100 hPa (which yields a value for a of about 1.5). Using Eq. (11), we then get for the subcolumns A^* , B^* , C^* :

$$A^* = X \times (A+B+C) / (X+Y+Z^*)$$

$$B^* = Y \times (A+B+C) / (X+Y+Z^*) \quad (12)$$

$$C^* = Z^* \times (A+B+C) / (X+Y+Z^*)$$

Equation (12) again satisfies the demand that $A^* + B^* + C^*$ equals $A+B+C$. At the same time there is no dependency on the Logan (1999) O_3 column climatology values anymore, only on the relative scaling between the (extended) Logan (1999) climatology layers. Clearly, the assumption given by Eqs. (11) and (12) should be further examined in more detail. Applying this approach should at least result in TOR which will deviate more from the Logan (1999) climatology and are more influenced by the TOMS and SBUV satellite measurements. It is also important to note that the TOR depends strongly on the actual tropopause height. Therefore, tropopause height changes should always be considered in the interpretation of the TOR data.

6. Actual variability of the total stratospheric O_3 column

One basic assumption of the retrieval method in Fishman et al. (2003) is that the total stratospheric O_3 column (SOC) does not vary much within a 5-day period. Typical TOR values range from about 10 DU for the remote equatorial Pacific to about 50 DU for continental mid-latitude regions. Thus, for this assumption to introduce a small error on the TOR the 5-day variability of the SOC should be less than about 5–10 DU

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at maximum, and preferably smaller. To investigate this hypothesis we first examine the total O₃ column over Samoa as measured by the TOMS satellite. Samoa is considered a clean Pacific equatorial location where tropospheric O₃ concentrations are generally very low throughout the troposphere, as it is remote from any major pollution source.

Furthermore, changes in the tropical tropopause height are generally small (see also Fig. 3). As a result, the tropospheric O₃ column is low (<15 DU according to Fishman et al., 2003), and variations between the seasons are small.

Figure 4a shows the total O₃ column over Samoa measured by TOMS (version 7). The seasonal cycle can easily be discerned, with higher column densities towards the end of the year. However, superimposed on the seasonal cycle is a highly varying signal. Figure 4b shows the 5-day standard deviation values of the total O₃ column for Samoa. Again, there is a seasonal cycle, with a higher variability during the season when the overall concentrations are higher. On average, the 5-day standard deviation of the total O₃ column is about 5 DU (similar to results from Ziemke et al. (1998), who, based on Microwave Limb Sounder stratospheric O₃ profile measurements, derived a typical 2σ variation of the tropical stratospheric O₃ column of 10 DU). However, this value varies between 1 and 15 DU, and thus can be as high as the total tropospheric O₃ column at Samoa. Since the variability of tropospheric total O₃ column is small for this remote site, the high variability of the total O₃ column densities must be caused by variations in the stratospheric O₃ column densities.

A similar picture occurs when we investigate the total O₃ column variability at Lauder (New Zealand). Lauder is a mid-latitude station, so that the seasonal cycle of the total O₃ column is much larger than that at Samoa. According to Fig. 5a, the total O₃ column densities at Lauder range from about 250 to 400 DU. The 5-day standard variation of the total O₃ column (Fig. 5b) is on average approximately 20 DU, with a range from 5 to 50 DU.

To further investigate the O₃ column variability we looked at stratospheric O₃ profiles obtained from lidar measurements at the site (Brinksma et al., 2002). There are typically about 100 profiles per year (about one every 3 days). Because of the large

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amount of O₃ profiles, we can compare the variability of the lidar profiles with the variability of the TOMS total O₃ column measurements. We took one year (1997) of data to investigate the variability. Figure 6 shows the stratospheric O₃ column variations for different layers (11–45, 15–45 and 19–45 km altitude layers) and the corresponding TOMS measurements. The O₃ column of above 45 km altitude is typically only 2 DU, and variations are small. We can assume that O₃ above 45 km adds little to the total O₃ column variability. The standard deviation of the TOMS measurements for this period is 32.4 DU for TOMS collocations with lidar measurements (31.6 DU for all TOMS measurements). The standard deviation for the lidar measurements are 26.4, 22.7 and 15.5 DU for O₃ columns between 11–45, 15–45 and 19–45 km, respectively. The variability of the O₃ column increases with decreasing altitude. This should not be a surprise considering that the upper-troposphere/lower stratosphere region is a much more dynamically active region than the middle and upper stratosphere. Lauder is a mid-latitude site, so that the tropopause generally will be below 15 km altitude, and during winter closer to 10 km altitude. About 2/3 of the total O₃ column variability occurs above 15 km altitude and can be considered stratospheric. What is more, the value of the standard deviation above 15 km altitude was 22.7 DU, which means that variations in the stratospheric O₃ column can be as high as 45 DU (2 times the standard deviation). This value is in agreement with the 5-day standard deviations, which, on occasion, could be as high as 50 DU. From this analysis we conclude that a constant stratospheric O₃ column densities for a 5-day period is not a reasonable assumption.

At the same time it must be mentioned that daily stratospheric O₃ column variations appear to occur randomly. In the TOR method of Fishman et al. (2003) the once-per-5-days SBUV total O₃ column measurements are subtracted from the every day total O₃ column measurements from TOMS. Over longer periods of time a random signal will average out, and it is likely that this is also the case for the TOMS minus SBUV total O₃ column densities. To estimate this effect we calculated the differences between the daily TOMS total O₃ column measurements and the corresponding 5-day TOMS total O₃ columns (from –2 days to +2 days; for the period 1996–2003). The

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average absolute difference for a 5-day period was 15.1 DU for Lauder and 4.0 DU for Samoa. For one month the values were 5.7 and 1.5 DU, and for three months the differences were 3.2 and 0.6 DU for Lauder and Samoa, respectively. As expected, the differences (“errors”) become less important when averaging over longer periods of time. However, for a single month the 5-day sampling of stratospheric O₃ columns introduces on average an error in the TOR of about 10–20% for Lauder and Samoa. Although these are only rough estimates of the error introduced by the 5-day sampling, they indicate that one should be careful when for example looking at specific single months. Furthermore, these examples show that further investigation is needed to assess the error due to stratospheric O₃ variability.

7. Summary and discussion

In this article we have shown that the method used to retrieve TOR columns as described in Fishman et al. (2003) and applied in Pierce et al. (2003) yields a TOR that can be considered as the O₃ climatology from Logan (1999) below the tropopause, superimposed with differences between every-day TOMS and once-per-5 days SBUV total O₃ column measurements. The TOR product therefore is strongly biased to the effect of tropopause height variations and only marginally relies on the used satellite data. We have shown that, by simply applying a realistic tropopause height estimation to the Logan (1999) climatology we can derive a “TOR” that is very similar to the TOR climatology presented by Fishman et al. (2003). As a result, we think that the interpretation given to TOR in the application presented in Fishman et al. (2003) is not correct. We have shown that the high TOR columns during summer over India and China are not the result of pollution but of a large increase in tropopause heights in this region. We believe that the formulation of the method as presented in Fishman et al. (2003) is probably not the method they had in mind. We have therefore made a possible suggestion to improve the TOR method using the basic assumptions as presented by the authors in an earlier publication (Fishman and Balok, 1999). However,

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we have also shown that the implicit assumption that the total stratospheric O₃ column is nearly constant over 5 days is not valid. This could pose another complication in this TOR methodology. On the other hand, the 5-day variability of the stratospheric O₃ column may occur randomly. It is possible that the error introduced by this assumption will average out when TOR columns are averaged over longer periods of time (months-seasons). Investigating TOMS total O₃ columns for Samoa and Lauder confirms that this effect becomes smaller when averaging over longer periods of time. At the same time, however, it shows that the errors are not that small that they can be ignored. We therefore conclude that the effect of stratospheric O₃ variability in combination with 5-day SBUV sampling on the final TOR column should be investigated in more detail.

Even though the method applied by Fishman et al. (2003) to retrieve TOR columns from satellite measurements is incorrect, the results of the retrieval do not appear to be unreasonable. As was shown, the Fishman TOR column is largely dependent on the Logan (1999) climatology. This climatology itself is based on a large number of O₃ sonde measurements. Therefore, the climatology is not an unreasonable approximation of the actual O₃ concentrations. And averaged over longer periods of time, the actual O₃ concentrations will not differ too much from the climatology. Furthermore, by scaling the Logan (1999) climatology to the actual tropopause height, the TOR column will also correlate well with synoptical systems and with seasonal variations in parameters since the tropopause height also changes with season. As a result, the TOR column variations appear to be very realistic. But it should be realized that for this satellite data is not needed.

In this paper we present an alternative to extend the Logan (1999) climatology to 63 hPa and to redistribute the O₃ in the three lowest SBUV layers. Further study is needed to optimize the Logan (1999) climatology extension. If the TOR product is being improved along the suggested changes, and it can be shown that the stratospheric variability does not introduce large errors in the TOR columns, the TOR columns could become a valuable dataset. However, until these changes have been applied, and a better validation of the dataset against independent measurements has been per-

formed, there is little new added information in the current dataset.

Moreover, one should be careful with interpretation of tropospheric O₃ columns. As was shown for India and China, the tropospheric O₃ columns are strongly dependent on tropopause height variations. The effects of tropopause height changes on total tropospheric columns obscures the observation of pollution, therefore the interpretation of tropospheric columns should always be accompanied by a description of the tropopause height changes, especially outside the tropics. Without this knowledge, it is not possible to fully understand the tropospheric O₃ column variability, which in turn can lead to wrong conclusions about the temporal and spatial variability of tropospheric O₃.

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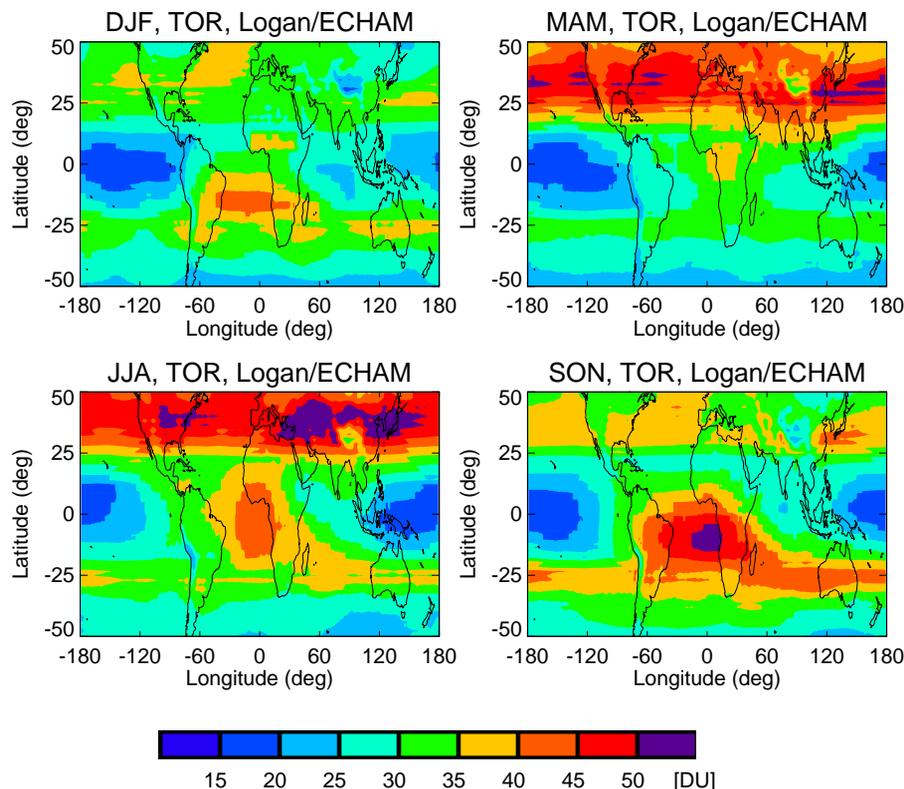
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Fig. 1. Mean seasonal total tropospheric O₃ columns (Dobson Units: 1 DU = $2.69 \cdot 10^{16}$ molecules cm^{-2}) using the Logan (1999) climatology and ECHAM tropopause height and surface elevation (DJF: December-January-February, MAM: March-April-May; JJA: June-July-August, SON: September-October-November). ECHAM horizontal resolution is approximately $1.9^\circ \times 1.9^\circ$ (T63). Model simulation was done for 1996.

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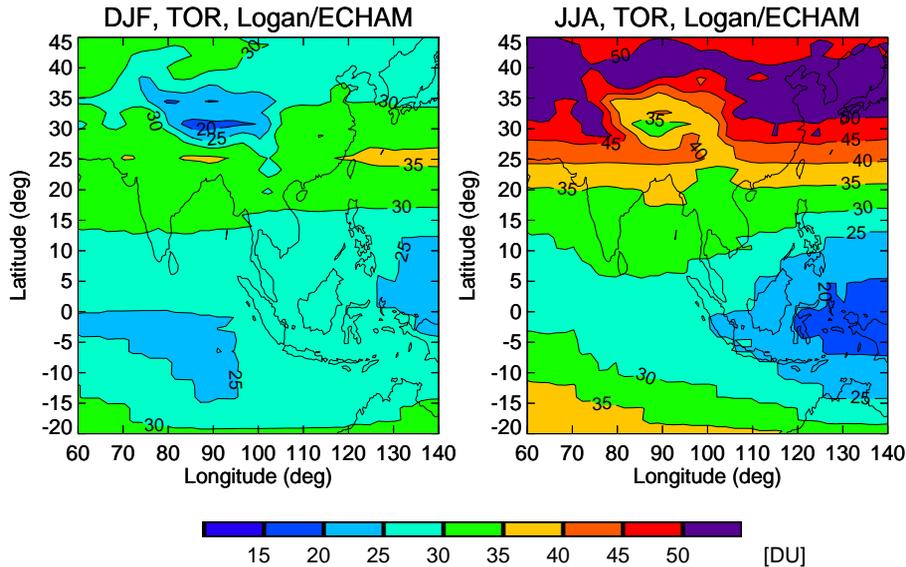


Fig. 2. As Fig. 1 but for southeast Asia for the DJF and JJA seasons.

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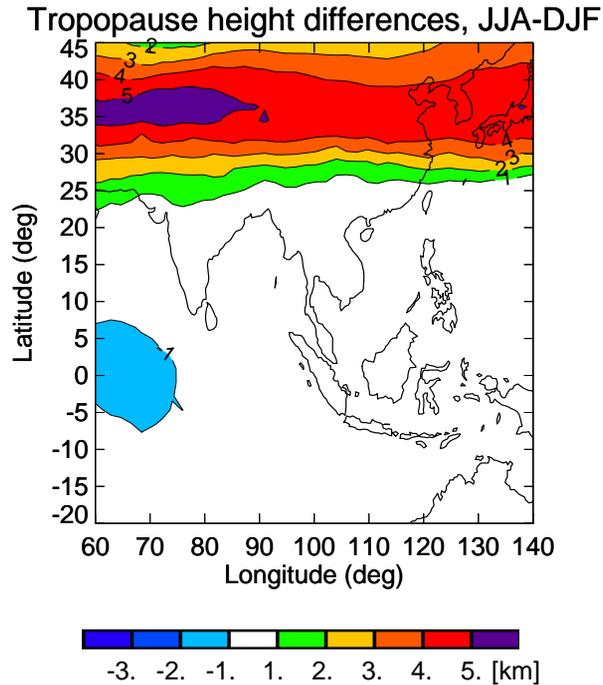
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Fig. 3. Mean tropopause height differences (km) between the seasons DJF and JJA derived from the ECHAM model simulation for 1996.

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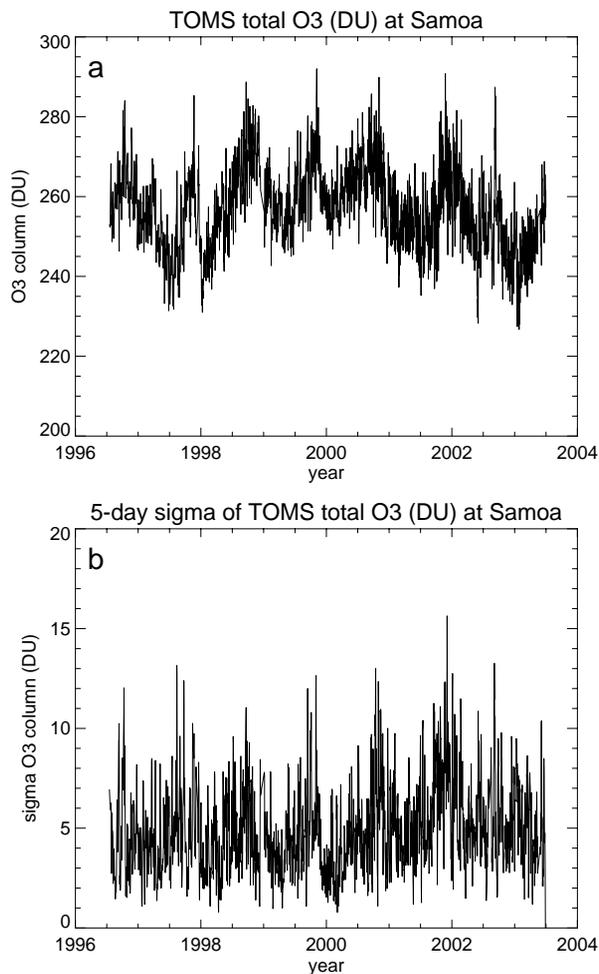
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Fig. 4. TOMS total O₃ column (DU) measurements over Samoa (14°15' S, 170°34' W) for the period 1996–2004, and the corresponding five-day standard deviation of the total O₃ columns.

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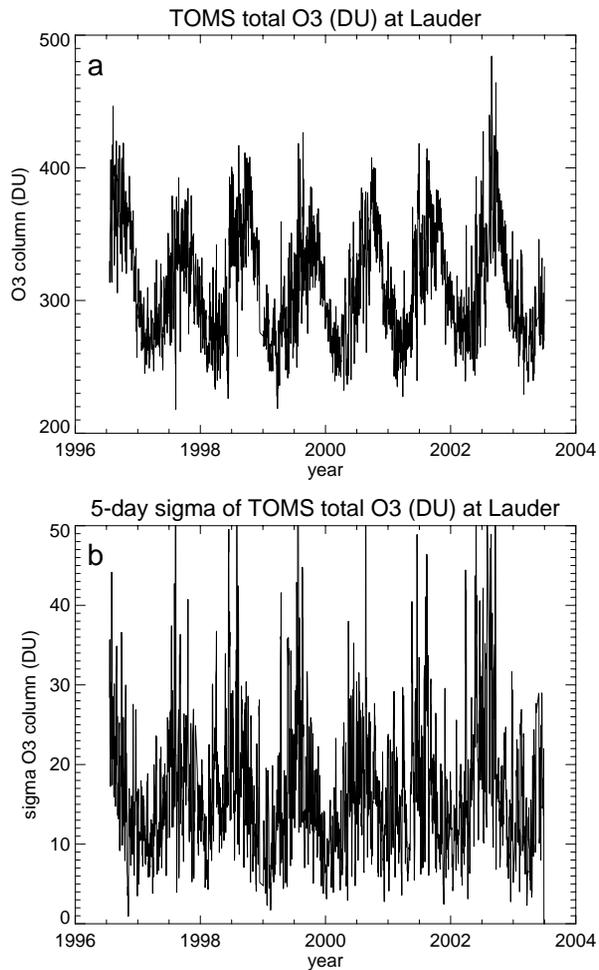
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Fig. 5. As Fig. 4 but for Lauder, New Zealand (45°02' S, 169°40' W).

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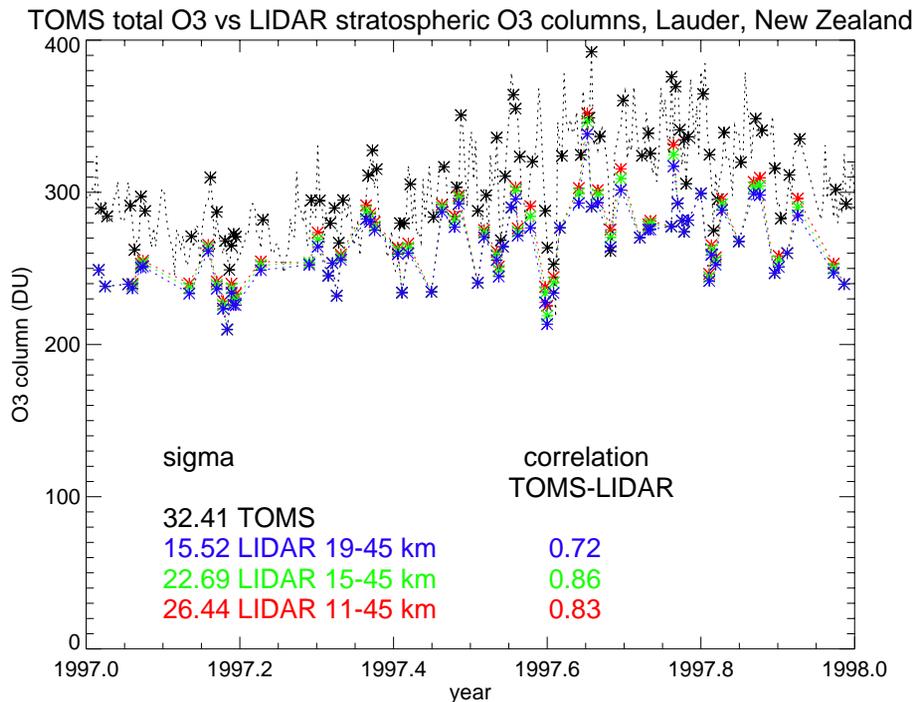


Fig. 6. O₃ column measurements (DU) over Lauder, New Zealand for 1997. O₃ columns from lidar measurements were calculated for the altitude ranges 11–45 km (red line), 15–45 km (green line) and 19–45 km (blue line). Total O₃ column measurements from TOMS are denoted by the black dotted line, with the collocations with lidar measurements indicated by the asterixes. Indicated are also the values of standard deviation of the lidar and TOMS measurements, and the correlations between the lidar O₃ columns and TOMS total O₃ column measurements.

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