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Analysis of Tropical Tropospheric Ozone Derivation from Residual-Type Method

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Abstract : During the northern burning season, biomass burning is found north of the equator, while satellite estimates from the residual-type method such as the CCD method show higher ozone south of the equator. This discrepancy is called the tropical Atlantic paradox (Thompson *et al.*, 2000). We use satellite and ground-based measurements to investigate the paradox. When the background tropospheric ozone over the Pacific Ocean from TOMS measurements is subtracted from the latitudinal total ozone distribution (e.g. TOMS-Pacific method), the results show remarkable agreement with the latitudinal stratospheric ozone distribution using the CCD method. The latitudinal tropospheric ozone distribution using the CCD method, with a persistent maximum over the southern tropical Atlantic, is also seen in the latitudinal tropospheric ozone distribution using the TOMS-Pacific method. It suggests that the complicated CCD method can be replaced by the simple TOMS-Pacific method. However, the tropical Atlantic paradox exists in the results of both the CCD and TOMS-Pacific methods during the northern burning season. In order to investigate this paradox, we compare the latitudinal ozone distributions using the CCD and TOMS-Pacific methods by using the SAGE measurements (e.g. TOMS-SAGE method) and the SHADOZ ozonesoundings (e.g. TOMS-Sonde method) assuming zonally invariant stratospheric ozone, which is the same assumption as of the CCD method. During the northern burning season, the latitudinal distributions in the tropospheric ozone derived from the TOMS-SAGE and TOMS-Sonde methods show higher tropospheric ozone over the northern tropical Atlantic than the southern Atlantic due to a stronger gradient in stratospheric ozone relative to that from the CCD and TOMS-Pacific methods. This indicates that the latitudinal tropospheric ozone distribution can be changed depending on the data that is used to determine the latitudinal stratospheric ozone distribution. Therefore, there is a possibility that the north-south gradient in stratospheric ozone over the Atlantic can be a solution of the paradox.

Key Words : Tropical Tropospheric Ozone, TOMS, Residual-type Method, Biomass Burning, Atlantic Paradox.

1. Introduction

Tropical tropospheric ozone plays a key role in both tropospheric oxidation and global climate change. It has been measured by ozonesondes for several decades, but the small number of locations for these observations and occasional aircraft campaigns provide a limited spatial coverage. Therefore, it is

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necessary to use the satellite-based method for retrieving tropical tropospheric ozone that provides a sufficient spatial coverage. In a study by Fishman and Larsen (1987), the concept of determining tropical tropospheric ozone by the Tropospheric Ozone Residual (TOR) method using satellite measurements was first utilized. This approach derived tropospheric ozone by subtracting stratospheric ozone taken from Stratospheric Aerosol and Gas Experiment (SAGE) from total ozone taken from Total Ozone Mapping Spectrometer (TOMS). Subsequent studies calculated tropical tropospheric ozone using the residual-type methods, which are the Convective Cloud Differential (CCD) method and the modified residual method (Hudson and Thompson, 1998; Ziemke *et al.*, 1998; Thompson and Hudson, 1999).

There is an argument that the latitudinal distribution of tropical tropospheric ozone using the residual-type methods differ from that using models over the Atlantic during boreal winter and spring. All satellite-based methods except the Scan-Angle Method (SAM) assume zonally invariant stratospheric ozone and always show a maximum over the southern tropical regions. However, SAM and the Geophysical Fluid Dynamics Laboratory Global Chemical Transport Model (GFDL/GCTM) derive a maximum over southern tropical regions during boreal summer and autumn, and northern tropical regions during boreal winter and spring, so that the individual maximums correspond to the biomass burning seasons in each tropical hemisphere (Galanter *et al.*, 2000; Kim *et al.*, 2005). This discrepancy is called the tropical Atlantic paradox (Thompson *et al.*, 2000). They suggested that ozone enhancement over the South Atlantic resulted from a combination of interhemispheric transport, aged stratospheric tropospheric air, and possibly from ozone supplied by lightning NOx. However, they noted that the appearance of the ozone was difficult to interpret with their limited shipboard measurements, 9 ozonesoundings between 15°S and 15°N.

In order to explore the paradox on the basis of the work of Thompson *et al.* (2000), most of studies have tried to interpret in-situ measurements with the limited ozonesondes data over the Atlantic (Edwards *et al.*, 2003; Jenkins *et al.*, 2003; Chatfield *et al.*, 2004). However, no one has ever focused on the assumption of the residual-type methods and investigated how a small deviation in the zonally-flat stratospheric ozone influences the latitudinal stratospheric ozone distribution and latitudinal tropospheric ozone distribution over the Atlantic. The purpose of this paper is to investigate how the latitudinal stratospheric ozone distribution influences the latitudinal tropospheric ozone distribution over the Atlantic. To accomplish this, we introduce three new simple tropospheric ozone retrieval methods using TOMS, SAGE, and ozonesonde measurements, based on the assumption of previous residual-type methods but relatively simple process to the previous residual-type methods using the complicated processes (Table 1). We will compare the CCD-derived tropospheric ozone distribution and stratospheric ozone with other methods. In addition,

Table 1. Characteristic features of the CCD, TOMS-Pacific, TOMS-SAGE, and TOMS-Sonde methods.

	CCD	TOMS-Pacific	TOMS-SAGE	TOMS-Sonde
Assumption	Zonally-flat stratospheric ozone			
Total ozone	TOMS total ozone			
Stratospheric ozone	TOMS ozone above the cloud tops near the tropopause	TOMS ozone over the Pacific	SAGE stratospheric ozone	Ozone subtracted Sonde tropospheric ozone from TOMS
Process	Complicacy	Simplicity	Simplicity	Simplicity

we will seek a possible linkage between the latitudinal stratospheric ozone distribution and the paradox.

2. Methodology

1) Convective Cloud Differential Method

From the analysis of SAGE, Halogen Occultation Experiment (HALOE), and Microwave Limb Sounder (MLS), Ziemke *et al.* (1998) drew a conclusion that the zonal variation of stratospheric ozone in the tropics is flat. Based on this assumption, the CCD method requires preliminary work in order to determine the latitudinal distribution of stratospheric ozone, which is the same as TOMS ozone above the cloud tops near the tropopause. After examining the high convective regions using International Satellite Cloud Climatology Project (ISCCP) cloud top pressures and a sophisticated statistical scheme, the latitudinal stratospheric column ozone is derived by averaging total ozone measured over the tropical Pacific Ocean between 120°E and 180°E. Then, the CCD method determines the tropospheric ozone by subtracting the zonally invariant stratospheric ozone from TOMS total ozone when the skies are clear. This implies that if any method measures the climatological latitudinal stratospheric ozone distribution for a single longitude band, the entire amount of tropical stratospheric ozone can be determined because of the zonal mean assumption. The tropospheric ozone can also determine by subtracting the latitudinal stratospheric ozone distribution from the TOMS total ozone distribution.

2) TOMS-Pacific Method

To determine the total amount of ozone TOMS combines the amount of ozone in the stratosphere and in the troposphere. The TOMS algorithm efficiently retrieves the amount in the stratosphere but less efficiently in the troposphere (McPeters *et al.*, 1996). This implies that the TOMS algorithm is less sensitive to tropospheric ozone information. Therefore, if the amount of tropospheric ozone varies only slightly over a spatial domain, any variation in the total ozone will arise from variations in stratospheric ozone because the algorithm would not have recognized any differences in the troposphere (Hudson *et al.*, 1995; Kim *et al.*, 1996). We have applied this physical concept to the longitudinal TOMS Version 8 Level 2 total ozone distribution over two selected regions: the Pacific Ocean with clean environment between 120°E and 180°E and the Atlantic between 15°W and 15°E with elevated tropospheric ozone from biomass burning. Fig. 1 shows the latitudinal distribution of TOMS total ozone subtracted by 26 DU with reflectivity less than 20% (cloud free conditions) over the Atlantic and the Pacific Ocean, and the latitudinal stratospheric ozone distribution from the CCD method for the TOMS Nimbus-7 (N7) period. The 26 DU is the suggested average background tropospheric ozone amount in the tropics by Hudson and Thompson (1998) and Kim *et al.* (1996). The latitudinal resolution of 5 degree is used to make it suitable for the CCD resolution. The latitudinal stratospheric ozone distribution from the CCD is quite different from that over the Atlantic because of the strong variability in the latitudinal tropospheric ozone distribution as seen from the satellite measurements. On the contrary, the latitudinal total ozone distribution subtracted by 26 DU over the Pacific Ocean (hereafter the "TOMS-Pacific" method) agrees remarkably well with the latitudinal stratospheric ozone distribution from the CCD. This agreement suggests that the latitudinal stratospheric ozone variation in the tropics is simply due to the lack of influence that anthropogenic activity has on the latitudinal total ozone variation over the Pacific Ocean.

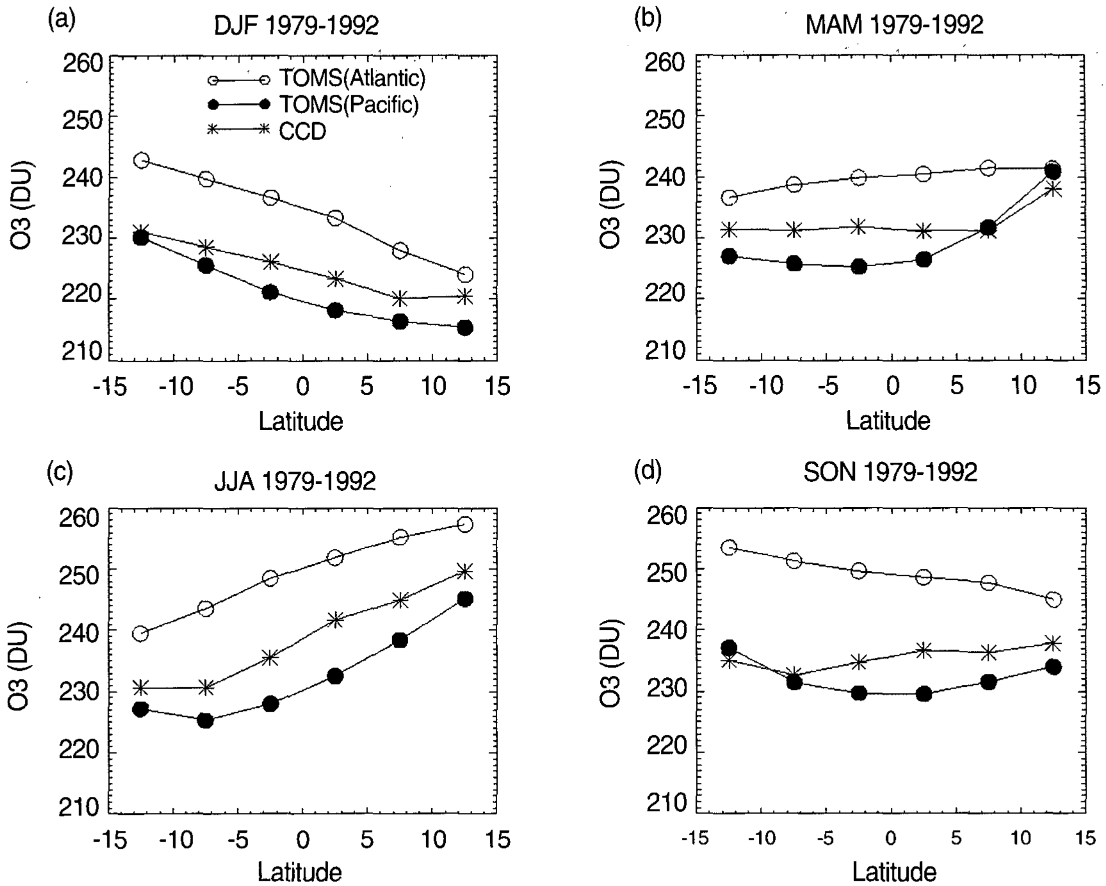

Fig. 1. The latitudinal distribution of TOMS total ozone over the Atlantic (open circles), and over the Pacific (closed circles), CCD stratospheric ozone (asterisks) averaged for 1972-1992 (TOMS/N7 period) in the (a) December-January-February (DJF), (b) March-April-May (MAM), (c) June-July-August (JJA), and (d) September-October-November (SON) time periods.

3) TOMS-SAGE method

We have used the improved SAGE-II V6.1 measurements (Wang *et al.*, 2002) averaged over 5 degree latitude bin based on the assumption of zonally invariant stratospheric ozone. Tropospheric ozone is then derived by subtracting averaged stratospheric ozone measured by SAGE from total ozone measured by TOMS. We define this technique as the TOMS-SAGE method.

4) TOMS-Sonde method

The Southern Hemisphere Additional Ozonesonde (SHADOZ) campaign has enough well distributed ozonesonde stations to determine the distribution of latitudinal ozone from the soundings. We have selected eight available ozonesonde stations in the tropics: Samoa(14°S, 171°W), Ascension Island(8°S, 14°W), Natal(5°S, 35°W), San Cristobal(1°S, 90°W), Nairobi(1°S, 37°E), Java(8°S, 113°E), Paramaribo (6°N, 55°W), and Hilo(19°N, 155°W). Thompson *et al.* (2003) examined the distribution of longitudinal tropospheric ozone by using SHADOZ measurements that were arranged longitudinally at given latitude. We used this concept for the distribution of latitudinal ozone from eight SHADOZ stations: 1) the tropospheric column ozone was determined by integrating the measurements from the surface to the tropopause as defined by WMO (1957) from the soundings and then subtracting it from the total amount of ozone measured by TOMS at each station to determine the stratospheric column ozone; 2) using the assumption of zonally invariant stratospheric ozone, averaging and then interpolating these data resulted in a latitudinal stratospheric ozone distribution for a 5-degree latitude bin. We define this as the TOMS-Sonde method. To be consistent with the ozonesonde data, which starts in 1998, we have used the products from CCD, TOMS-Pacific, TOMS-SAGE, and TOMS-Sonde techniques for 1998-2000.

3. Results

1) Spatial distribution

Fig. 2 shows the spatial distribution of tropospheric ozone derived from the CCD, the TOMS-Pacific, the TOMS-SAGE, and the TOMS-Sonde for the DJF period. Tropospheric ozone derived from the CCD (Fig. 2a) has a noticeable enhancement over the southern Atlantic and a minimum over the central Pacific. This distribution is perfectly consistent with the distribution of the TOMS-Pacific, but about 4DU higher in maximum ozone amounts (Fig. 2b). However, the latitudinal distribution over the Atlantic of the CCD and the TOMS-Pacific is significantly different from that of TOMS-SAGE and TOMS-Sonde (Figs. 2c and d). The distribution of the enhanced ozone over northern equatorial Africa from the TOMS-SAGE and the TOMS-Sonde correlated well with the northern dry season, which is strongly influenced by biomass burning activity. This feature for the DJF period can be seen in the amount of tropical tropospheric ozone shown by SAM and model (Galanter *et al.*, 2000; Martin *et al.*, 2002; Kim *et al.*, 2005). We will explore the latitudinal discrepancy over the Atlantic in detail in the following section.

2) Latitudinal distribution

Fig. 3 shows the latitudinal distribution from the CCD, TOMS-Pacific, TOMS-SAGE, and TOMS-Sonde techniques for 1998-2000. During the DJF period (Fig. 3a) variations of less than 10 DU in the distribution of latitudinal stratospheric ozone measured by CCD and TOMS-Pacific are responsible for the latitudinal gradient of about 15 DU in total ozone, but that variations of 15 DU in the distribution of latitudinal stratospheric ozone measured by TOMS-SAGE and TOMS-Sonde are responsible for the total ozone gradient. In spite of the same negative north-south gradient for the DJF period, the latitudinal gradient in stratospheric ozone from the CCD and TOMS-Pacific methods is 5-9 DU smaller than that from the TOMS-SAGE and the TOMS-Sonde methods. Because the relatively lower amounts of stratospheric ozone in the southern tropical Atlantic is subtracted from the total amount of ozone determined by TOMS, the latitudinal tropospheric ozone derived from the CCD and the TOMS-Pacific present a peak over the southern tropics. On the other hand the latitudinal tropospheric ozone distributions from the TOMS-SAGE and the TOMS-Sonde exhibits a maximum over the northern tropical Atlantic. This implies that the north-south gradient in tropospheric ozone distribution can be reversed depending on what data is used to determine the stratospheric component.

When the north-south gradient in total ozone is relatively small, which is shown in the MAM period (Fig. 3b), the latitudinal stratospheric ozone distribution from TOMS-Sonde significantly differs from that from the CCD, the TOMS-Pacific, and the TOMS-SAGE. The TOMS-Sonde attributes the latitudinal total ozone variation during the MAM period over the Atlantic mostly to tropospheric ozone,

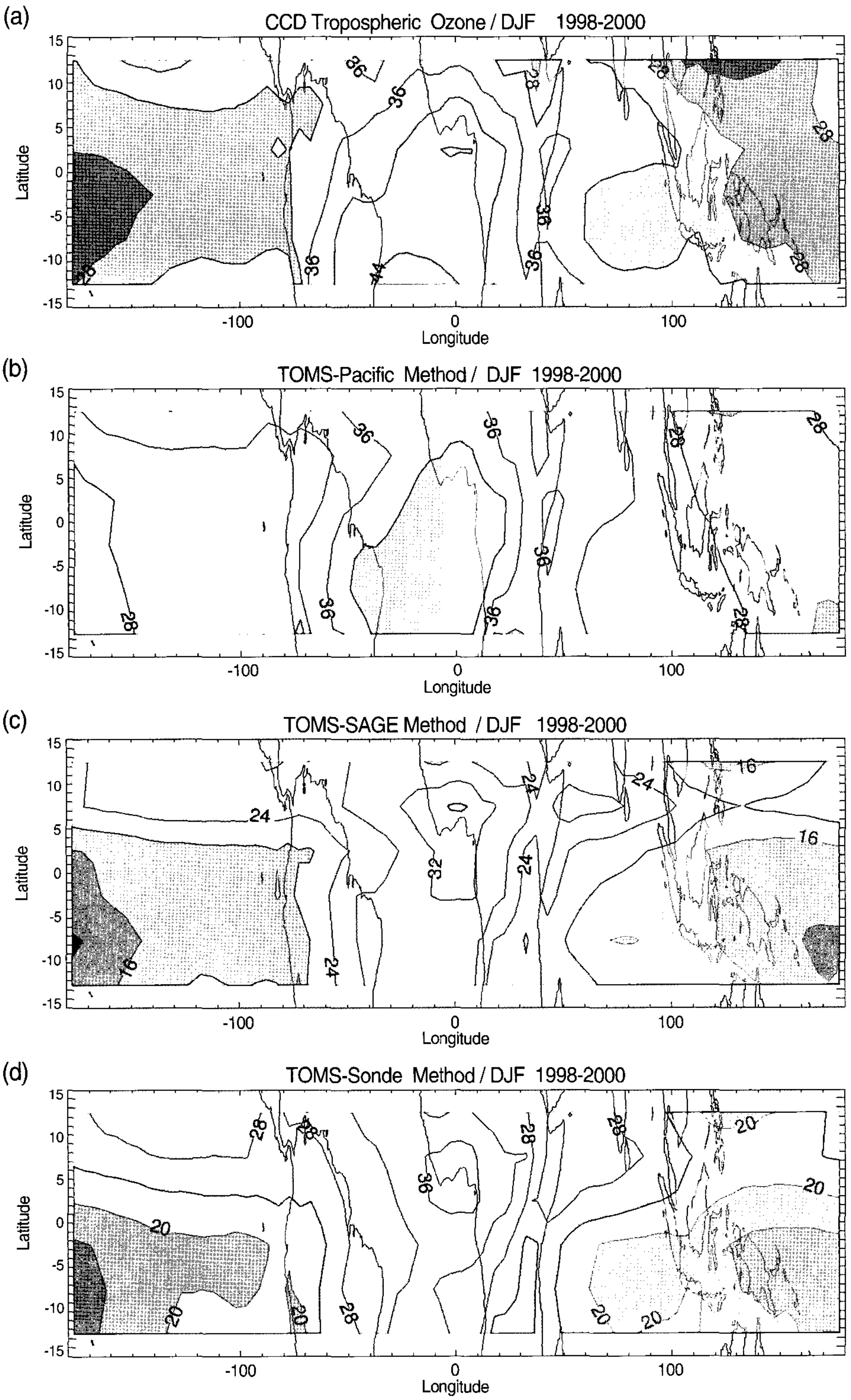

Fig. 2. The spatial distribution of tropospheric ozone from (a) the CCD method, (b) the TOMS-Pacific method, (c) the TOMS-SAGE method, and (d) the TOMS-Sonde method averaged from 1998-2000 in the December-January-February (DJF) time period

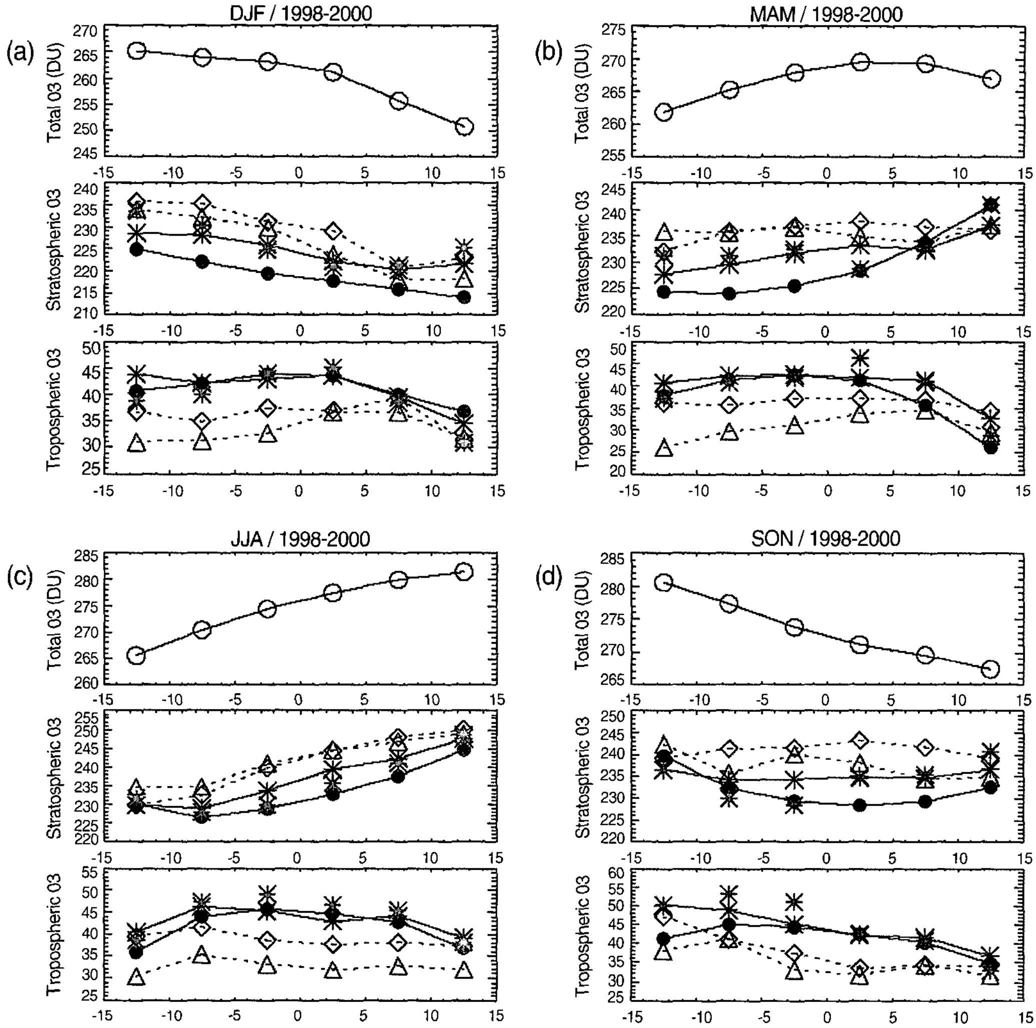

Fig. 3. The latitudinal distribution of total ozone (top panel), stratospheric ozone (middle panel), and tropospheric ozone (bottom panel) over the Atlantic Ocean between 15°W and 15°E for 1998-2000 in the (a) December-January-February (DJF), (b) March-April-May (MAM), (c) June-July-August (JJA), and (d) September-October-November (SON) time periods. The stratospheric and tropospheric ozone of 6 latitude bins calculated from the TOMS-SAGE

but the CCD, the TOMS-Pacifc, and the TOMS-SAGE attribute the variation mostly to stratospheric ozone. In addition, the CCD, the TOMS-Pacific, and the TOMS-SAGE show maximum tropospheric ozone over the south Atlantic, while the TOMS-Sonde shows a local maximum over the north Atlantic for the MAM period.

The latitudinal stratospheric ozone distribution from the CCD perfectly matches the distribution from the TOMS-Pacific in tendency and magnitude for the JJA period (Fig. 3c). The latitudinal stratospheric ozone distribution from the TOMS-SAGE and the TOMS-Sonde follows the same tendency as the CCD and the TOMS-Pacific, but at a magnitude of about 5-10 DU higher. Thereby, the north-south gradient in tropospheric ozone distribution from the TOMS-SAGE and the TOMS-Sonde is about 5-10 DU smaller than that from the CCD and the TOMS-Pacific.

The latitudinal stratospheric ozone distribution from the CCD and the TOMS-Pacific shows a local minimum near the equator for the SON period (Fig. 3d). However, the stratospheric ozone variation measured by the TOMS-SAGE and the TOMS-Sonde shows a local maximum near equator. The latitudinal tropospheric ozone distribution derived from all methods shows a southern maximum and northern minimum due to a strong north-south gradient in total ozone over the Atlantic. This feature is consistent with the satellite-based methods and model (Fishman *et al.*, 1990; Thompson and Hudson, 1999; Martin *et al.*, 2002; Kim *et al.*, 2005).

The latitudinal tropospheric ozone distributions from all methods detect increased ozone during the southern burning season. The tropospheric ozone from the TOMS-SAGE and TOMS-Sonde detect increased ozone during the northern burning season with a peak at 7.5°N, while the tropospheric ozone from the CCD and TOMS-Pacific show a broad enhancement over the equatorial south Atlantic. The CCD corrected for the TOMS error associated with clouds (Liu, 2003) intensifies the southern tropospheric enhancement during the southern burning season. However, the tropospheric ozone from the corrected CCD shows the peak moving northward during the northern burning season. It suggests that the paradox seen in the CCD can be due in part from the error in the TOMS data caused by cloud interference.

4. Conclusions

This study introduces new simple methods for retrieving tropical tropospheric ozone measurements assuming zonally invariant stratospheric ozone. In addition this study compares the CCD with TOMS-Pacific in the stratospheric and tropospheric distribution. Despite distinct differences in stratospheric ozone sampling, the latitudinal distribution of stratospheric ozone from the TOMS-Pacific method shows remarkable agreement with that from the CCD method. Therefore, the total amount of ozone determined by TOMS can simply be used for the latitudinal stratospheric ozone distribution instead of CCD stratospheric ozone, which requires a complicated process to compute. The latitudinal tropospheric ozone distribution from the CCD method, with a persistent maximum over the southern tropical Atlantic, is also seen in the latitudinal tropospheric ozone distribution from the TOMS-Pacific method. Therefore, the sophisticated CCD method can be replaced by the simple TOMS-Pacific method.

The longitudinal distribution of tropospheric ozone from the CCD, the TOMS-Pacific, the TOMS-SAGE, and the TOMS-Sonde exhibits a maximum over the Atlantic and a minimum over the Pacific. In addition, the CCD and the TOMS-Pacific always observes higher tropospheric ozone over the southern tropical Atlantic than over the northern. However, both the TOMS-SAGE and the TOMS-Sonde exhibit a maximum over the northern tropical Atlantic during the northern burning season and over the southern tropical Atlantic during the southern burning season, which is consistent with the oscillation of burning in the tropics. This latitudinal discrepancy is due to the difference in north-south gradient between stratospheric ozone from the CCD and the TOMS-Pacific and that from the TOMS-SAGE and the TOMS-Sonde. Therefore, that the north-south gradient in tropospheric ozone distribution can be reversed depending on what data is used to determine the stratospheric component.

A correction for the effect of clouds on stratospheric ozone determined by CCD changes the latitudinal tropospheric ozone distribution. Uncertainty in the amount of stratospheric ozone over the Atlantic can be an important cause of the paradox. Therefore, the residual-type method using reliable stratospheric ozone data will resolve the paradox.

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