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Ozone variation in the tropical tropopause layer as seen from ozonesonde data

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[1] Ozone variations with seasonal and intraseasonal timescales in the tropical tropopause layer (TTL) are investigated using a 5-year tropical ozonesonde data set from the SHADOZ (Southern Hemisphere Additional Ozonesondes) archive. The longitudinal ozone distribution in the tropical upper troposphere (TUT) shows a zonal wave one structure with maxima around the Atlantic and Africa and minima around the western Pacific throughout the year, while the annual variation shows maxima during northern summer to autumn at most longitudes. We compare the ozone distribution with the vertical temperature structure and found that the lapse rate is gradual (steep) at the ozone-enhanced (reduced) longitude and season. The east-west temperature structure and ozone variation in the TUT may be explained by the longitudinal variation of the large-scale atmospheric responses to the tropical heat source, which could govern both the temperature structure and the vertical transport processes. Ozone variability in the TUT is also large around the Atlantic and Africa and small around the western Pacific. However, the zonal wave one structure is not clear in the temperature variability and in the correlation coefficient between ozone and temperature, which can be related with wave activities around the tropopause. Remarkably large ozone variabilities with good correlation are observed in Africa during summer and in the central Pacific during autumn-winter. These are associated with large-scale equatorial waves, but the longitudinal variation of the wave activities does not seem to be an important factor in the zonal wave one structure of ozone.

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1. Introduction

[2] The tropical upper troposphere (TUT) and tropical lower stratosphere (TLS) is an important region from the viewpoint of stratosphere-troposphere exchange (STE), because the TUT/TLS is a passage from the troposphere to the stratosphere for mass and chemical species which affect stratospheric air conditions. In recent years, the TUT/TLS region has been considered as a transition layer between the troposphere and the stratosphere. So far, there are many definitions of the tropical tropopause corresponding to different aspects, such as thermal structure, convection, radiation, and chemical species. Recently, these concepts have been combined as one finite transition layer, the tropical tropopause layer (TTL), usually located around 12–19 km [e.g., *Highwood and Hoskins*, 1998], although an idea of the transition layer is not new [e.g., *Atticks and Robinson*, 1983]. To understand the STE processes in the TTL more quantitatively, a number of studies have focused on shorter timescale processes, such as horizontal wind circulations [e.g., *Pfister et al.*, 2001; *Holton and Gettelman*, 2001; *Hatsushika and Yamazaki*, 2003], convection [e.g., *Danielsen*, 1982; *Sherwood and Dessler*, 2001], equatorial waves [e.g., *Fujiwara et al.*, 2001], and eastward-propagating systems [*Eguchi and Shiotani*, 2004], in addition to a slow ascent [e.g., *Holton et al.*, 1995].

[3] Trace gases such as ozone are useful for understanding the STE process, because the photochemical lifetime in the TTL is much longer than the timescale of our interest described above. There are a few studies focusing on the relationship between vertical ozone distribution and the STE processes, though the seasonal and longitudinal variations of ozone in the TTL are still not clear owing to sparseness of tropical regular soundings with high vertical resolution. On the basis of the ozonesonde data at American Samoa in the central Pacific, *Folkins et al.* [1999] showed a gradual increase of ozone around 14 km, a few kilometers below the traditional thermal tropopause which is defined by the World Meteorological Organization lapse rate criterion or the cold point tropopause (CPT). This is partly because most convective clouds cannot reach up to the thermal tropopause, but stall around 14 km where we usually see the neutral buoyancy level. A level of zero radiative heating rate in the clear-sky condition is also located around here [*Folkins et al.*, 1999; *Gettelman et al.*, 2004; *Folkins and Martin*, 2005]. Below this level, the ozone concentration could be almost constant because of

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convective mixing with low ozone in the marine boundary layer. The relation between convection and thermal structure is discussed by *Gettelman and Forster* [2002] as a bottom of the TTL. In addition, a rapid transport process of the low-ozone air mass in the marine boundary layer up to the TUT could result in near-zero ozone sometimes observed in the equatorial Pacific TUT [*Kley et al.*, 1996; *Solomon et al.*, 2005].

[4] Equatorial wave can also play an important role in the STE process in the TTL. *Fujiwara et al.* [1998] using ozonesonde observations in Indonesia during the northern spring, at the beginning of the local dry season, showed an episode of equatorial Kelvin wave breaking, resulting in vertical mixing or downward transport of stratospheric ozone-rich air to the TTL. However, the space-time variation of Kelvin wave activity and the quantitative evaluation of Kelvin wave breaking are not clear yet. The transport or mixing process between the TUT and the middle-latitude lower stratosphere associated with Rossby wave breaking might be also a possible ozone source in the TTL. Particularly in the central tropical Pacific and the tropical Atlantic during northern autumn to spring, it is known that the Rossby wave propagates in the westerly duct [*Waugh and Polvani*, 2000], but a quantitative estimate of the Rossby wave mixing is not clear either.

[5] The primary motivation of this paper is to investigate controlling factors in the zonal variation of ozone in the TUT. *Thompson et al.* [2003b] showed a longitude-altitude section of the ozone mixing ratio in the troposphere from tropical ozonesonde soundings [see *Thompson et al.*, 2003b, Figure 4]. Though they did not pay much attention to the ozone variations in the TTL, a zonal wave number one structure with maxima around the Atlantic and Africa and minima around the western Pacific in the TUT can be seen in the figure. The zonal wave one structure of ozone concentration has been first pointed out as a tropospheric phenomenon from satellite observations in the column value [e.g., *Fishman et al.*, 1990; *Shiotani and Hasebe*, 1994], and now, the vertical distribution is clearly observed using ozonesonde observations.

[6] To view this wave one structure, however, *Thompson et al.* [2003b] and also a recent study by *Sauvage et al.* [2006] using MOZAIC (measurement of ozone and water vapour by airbus in-service aircraft) data set mostly focused on tropospheric production and transport processes of ozone and did not pay much attention to ozone variability in the TUT. The vertical ozone distribution in the TUT can also be obtained from satellite observations [*Wang et al.*, 2006], but there are several limitations as follows: no data in cloudy areas, poor vertical resolution, and no simultaneous temperature measurement.

[7] Seasonal ozone variation in the TTL is another important point to discuss. It has been shown, for example, by *Oltmans et al.* [2001] over the equatorial Pacific and by *Thompson et al.* [2003b] mainly in the Atlantic and at Africa from ozonesonde data. However, they did not pay much attention to the zonal variations in the whole tropics especially around the TTL. They also did not focus on the temperate structure simultaneously in the TTL. There are some studies focusing on the zonal temperature structure in the TTL [e.g., *Randel et al.*, 2003], but they are not so interested in the ozone variation. Some papers focused on the vertical ozone variation with the temperature structure in the TTL [e.g., *Folkins et al.*, 1999, 2002], but they did not pay much attention to the longitudinal or seasonal variation, though these papers focused on both radiative and chemical effects in detail.

[8] Using a 5-year tropical ozonesonde data set with high vertical resolution, we will investigate the longitudinal and seasonal variations of ozone in the TUT by paying attention to the zonal wave one structure and compare with the temperature structure in the TUT. Though the ozone amount in the TUT may not be a primary contribution to the tropospheric column value, it is important to investigate the zonal variations in the TUT from the viewpoint of the STE processes. We also focus on the influence of equatorial wave activity to ozone distribution and variability around the tropopause. Data description is in section 2. Results of ozone variations are presented in section 3, and section 4 summarizes our results.

2. SHADOZ Ozonesonde Data Set

[9] We use the electrochemical concentration cell (ECC) ozonesonde data provided by the Southern Hemisphere Additional Ozonesondes (SHADOZ) project [*Thompson et al.*, 2003a]. Almost once-per-week observations at the maximum have been made from 1998 to present for ozone, temperature, pressure, and relative humidity. The primary scope of the SHADOZ project is to validate satellite observations by filling up the data sparse region of ozonesonde soundings especially in the tropics. Before the SHADOZ project, it was difficult to describe the longitudinal structure of the tropical ozone distribution with high vertical resolution, but we now have 13 regular ozone sounding stations in the tropics and subtropics. As described by *Johnson et al.* [2002], *Thompson et al.* [2003a], and *Thompson et al.* [2007], there are instrumental differences of the ECC ozonesondes and the solutions, but these effects are assumed to be small in the troposphere and the TTL.

[10] The data for about 5 years during mid-1998 to mid-2003 are used in this study. Because the tropical tropospheric temperature over the whole tropics is much warmer in early 1998 under the El-Niño condition, and large ozone enhancement was observed at some stations especially over Indonesia, we did not use the data for the period of early 1998. In this study, ozonesonde data at nine stations close to the equator are used (Figure 1 and Table 1). The vertical resolution is set to 50 m with a linear interpolation method. The accuracy of the ECC ozonesonde is typically 5% in the troposphere, but it could be getting worse at very low ozone condition in the TUT [e.g., *Oltmans et al.*, 2001].

3. Results

3.1. Ozone and Temperature Profiles

[11] As the annual cycle of the tropopause altitude is highest in northern winter and lowest in northern summer in the tropics [e.g., *Seidel et al.*, 2001], first, we focus on ozone profiles for two seasons in December–February (DJF) and June–August (JJA) with a mean value and one standard deviation (Figure 2). To see the gross feature in the TUT/TLS, we will chiefly show results at five stations out of nine as follows. Ascension in the Atlantic and Nairobi in Africa are located around the zonal maxima of the TUT

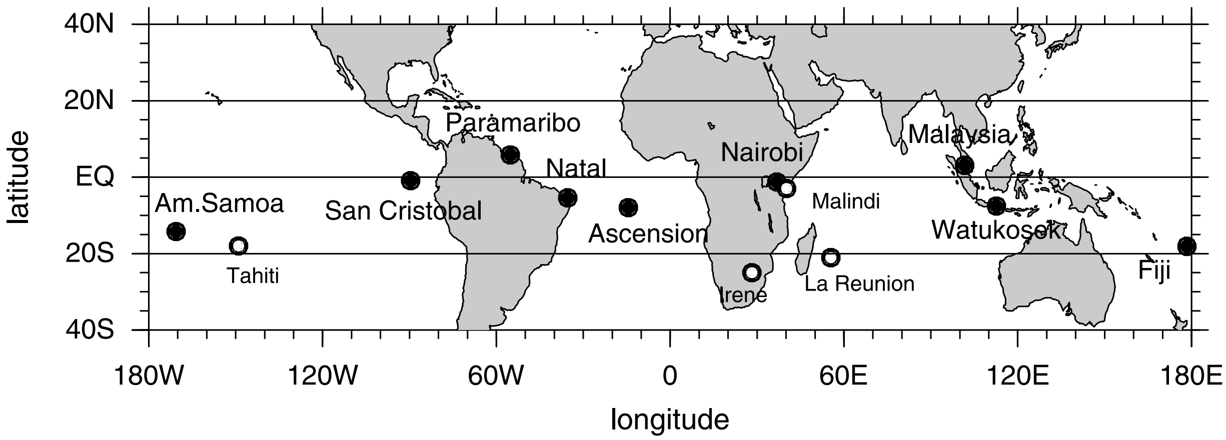

Figure 1. Locations of the tropical ozonesonde stations from SHADOZ (Southern Hemisphere Additional Ozonesondes) as of December 2003. Data at stations indicated by closed circles are used in this study.

ozone. Watukosek in Indonesia is located around the zonal minima. Fiji in the central Pacific and San Cristóbal in the eastern Pacific are located between the maxima and minima. Results for the five stations are set as in the leftmost frame at Ascension Island, where we see the zonal ozone maxima, and from the west to east stations.

[12] Comparing with the mean profiles over the whole tropics, the zonal wave one structure in the TUT with maxima around the Atlantic (Ascension) and Africa (Nairobi) and minima at the western Pacific (Watukosek) for both seasons is clear as described by *Thompson et al.* [2003b]. A gradual increase of ozone concentrations about 1–4 km below the tropopause is observed as well as a steep increase above the tropopause. The gradual increase of ozone at a few kilometers below the tropopause was pointed out by *Folkins et al.* [1999] from ozonesonde soundings at American Samoa in the central Pacific. A level where the ozone concentration gradually goes up to the stratospheric value is sometimes called the ozonopause [e.g., *Fujiwara et al.*, 2001]. It is usually located lower than the thermal tropopause, but difficult to define quantitatively. We set it as a level of 70-ppbv ozone concentration, and we call this point as the bottom of the ozone TTL. If we suppose the top of the ozone TTL as the cold point tropopause (CPT; hereafter we use the CPT as the tropopause), the thickness of the ozone TTL in DJF is ~5 km at Ascension, in contrast to ~1 km at Watukosek (see Figure 2).

[13] At Ascension, remarkably high ozone concentration is observed in nearly the whole troposphere with >50 ppbv of mixing ratio above 5 km in both seasons. The ozone concentration gradually increases with increasing altitude around 13–14 km about 3–4 km below the tropopause. At Nairobi we can also see ozone enhancement throughout the troposphere. The ozone concentration is large above 12 km with >50 ppbv in both seasons, and a steep increase of ozone is observed around 14 km in JJA. On the other hand, at Watukosek, the ozone concentration is nearly constant in almost whole of the troposphere with low mixing ratios ~25 ppbv, and a steep increase of ozone is observed just below the tropopause around 16 km in DJF. Fiji and San Cristóbal are located between the ozone maximum and minimum of the zonal wave one structure, and a gradual increase of ozone is observed around ~12–15 km.

[14] At most stations, ozone maxima in the middle troposphere (around 5–10 km) are often observed in the mean profiles, resulting from some ozone-rich layered structures in each profile. These maxima may be due to the air mass transport from continental sources [e.g., *Oltmans et al.*, 2001] and have been indicated especially in northern autumn around the Greenwich meridian by *Thompson et al.* [2003b], but they are clearly separated from the stratospheric ozone enhancement.

[15] At the ozone-enhanced longitudes (around the Greenwich meridian; Ascension and Nairobi), ozone variability is large in the middle and upper troposphere. In contrast, at the ozone minimum longitudes (around western Pacific; Watukosek), ozone variability is small even just below the tropopause. It is supposed that tropospheric air is well mixed and the ozone concentration is kept constant with a low value. At Fiji, ozone variability is large above 12 km with some ozone-enhanced (>100 ppbv) profiles in DJF, but the averaged value is not so large as that of Ascension. The averaged ozone value is greater than the mode, indicating its skewness in the distribution. Though the skewed distributions were also pointed out at some SHADOZ stations as a tropospheric phenomenon by *Thompson et al.* [2003b], the reason was not clearly described yet. It might be important that episodic wave events entrain ozone-rich air mass in the TUT. At San Cristóbal, the mean value is close to the mode.

Table 1. Summary of Ozonesonde Stations Used in This Study From the SHADOZ Archive

Station	Longitude	Latitude	Elevation, m
American Samoa	170.56°W	14.23°S	77
San Cristóbal (Galapagos)	89.60°W	0.92°S	8
Paramaribo, Suriname	55.21°W	5.81°N	25
Natal, Brazil	35.38°W	5.42°S	42
Ascension Island	14.42°W	7.98°S	91
Nairobi, Kenya	36.80°E	1.27°S	1795
Kuala Lumpur, Malaysia	101.7°E	2.73°N	17
Watukosek (Java), Indonesia	112.65°E	7.57°S	50
Fiji	178.40°E	18.13°S	6

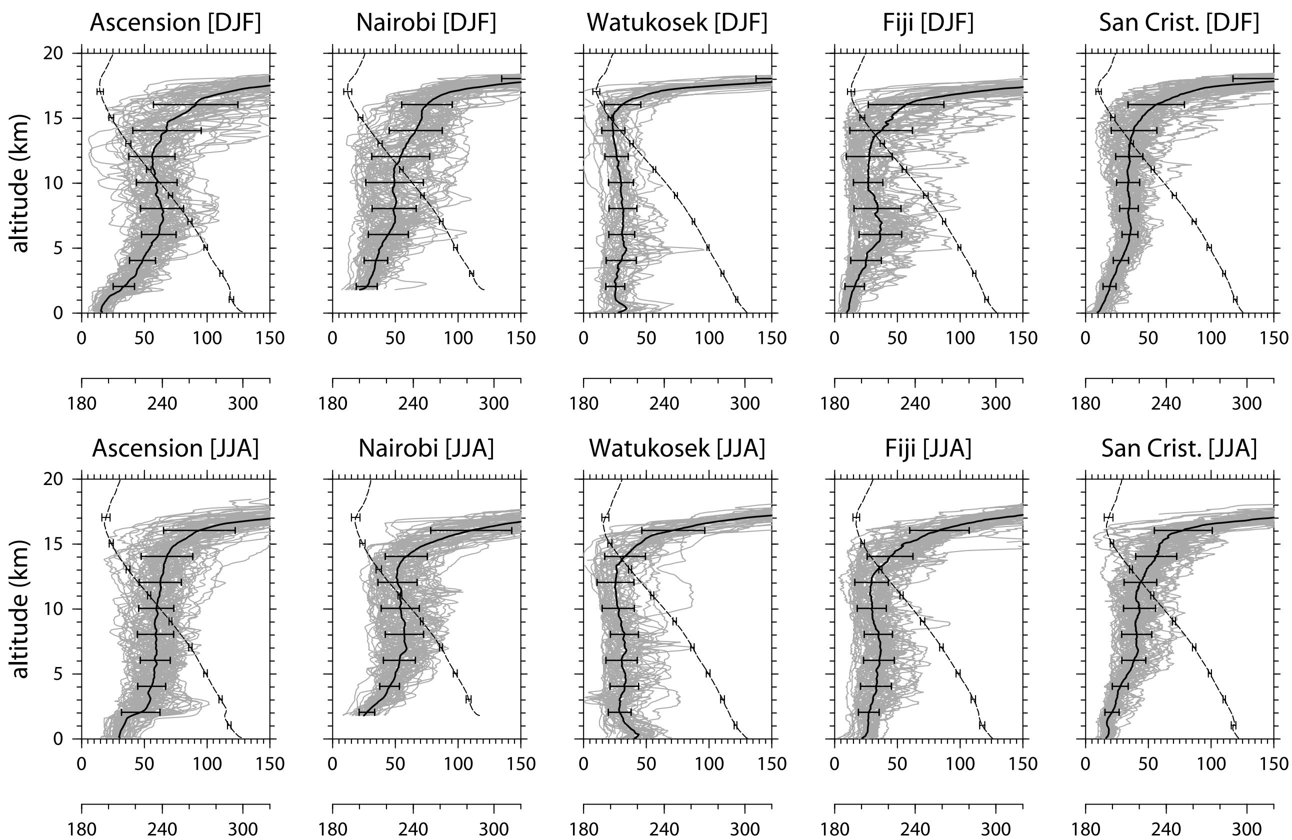

Figure 2. Vertical profiles of the ozone mixing ratio (solid line in parts per billion by volume; upper scale) and the averaged temperature (dashed line in kelvin; lower scale) for December–February (top) and June–August (bottom) at Ascension, Nairobi, Watukosek, Fiji, and San Cristóbal. Thick lines indicate averaged ozone profiles. Error bars indicate ±1 standard deviations.

[16] Comparing ozone profiles between northern winter and summer in the TUT, we can clearly see the seasonal variation at each station. For example, at Watukosek, a steep increase of ozone with small ozone variability is observed just below the tropopause in DJF (∼16 km), while a more gradual increase with larger variability is observed in JJA (around ∼13–15 km). Generally larger ozone mixing ratios and larger variabilities are observed in northern summer than in winter.

[17] To infer the relation between the ozone concentration and background thermal structure, temperature profiles for 10–20 km in DJF and JJA are shown in Figure 3. Though the tropopause altitude does not change so much in both seasons and at each station, there are longitudinal variations

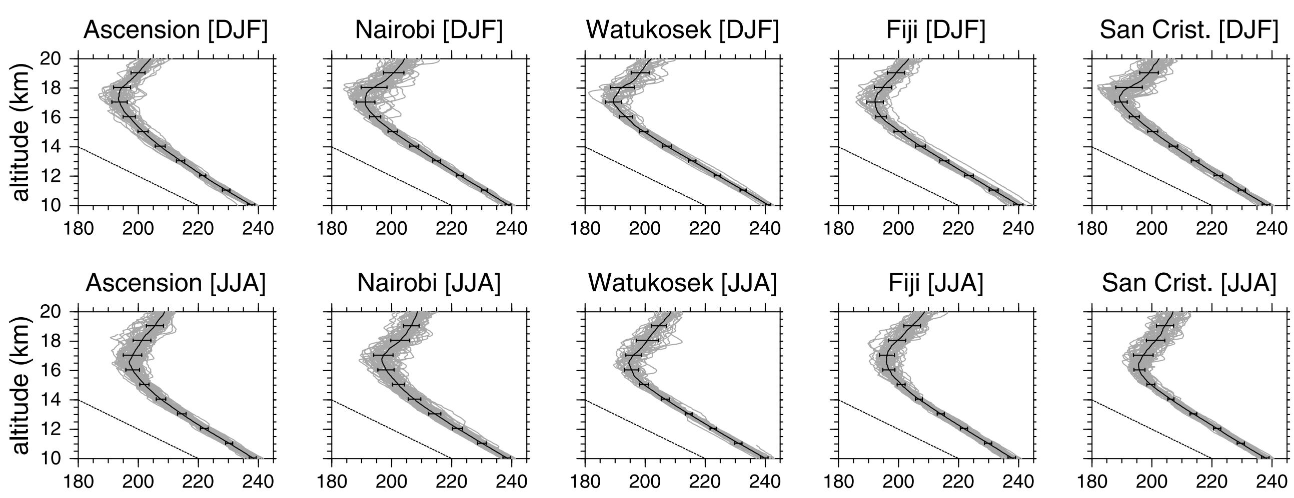

Figure 3. As Figure 2 but for the temperature for 10–20 km. Thick lines indicate averaged profiles, and error bars indicate ±1 standard deviations. Dashed lines indicate -10 K km^{-1}.

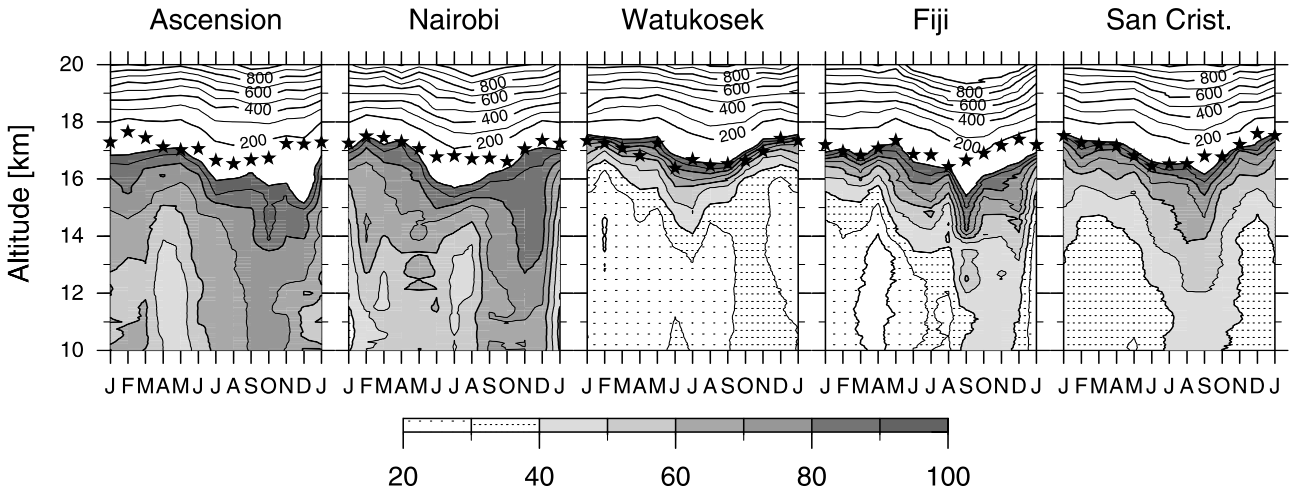

Figure 4. Time-altitude sections of the ozone mixing ratio (ppbv) at Ascension, Nairobi, Watukosek, Fiji, and San Cristóbal. The contour interval is 10 ppbv from 0 to 100 ppbv, and 100 ppbv from 100 to 1000 ppbv. Stars indicate locations of the cold point tropopause.

of the vertical temperature structure between ~12–16 km. The temperature gradient around 12–16 km seems to be weaker at the ozone-enhanced longitudes (Ascension and Nairobi) than at the ozone-poor longitudes (Watukosek). The vertical gradient of temperature is often called the lapse rate, and a weaker gradient corresponds to stable condition and vice versa. It should be noted that the lapse rate variations are clearly seen around 14 km, because this is a nodal height of the temperature variation in ozone-enhanced longitudes and seasons; there exist cold anomalies below 14 km and warm anomalies above 14 km, and vice versa. It might be difficult to recognize changes in the lapse rate in Figure 3 but the details will be seen in Figure 5. Comparing the two seasons, we generally observe a weaker gradient in JJA than in DJF, and remarkably weak gradient in the upper troposphere is observed at Nairobi in JJA. Temporal variation of the temperature variability in longitude or season is not clear in comparison with that of the ozone variability.

[18] From these ozone and temperature profiles, we can conclude qualitatively that the ozone distribution and the vertical temperature structure in the upper troposphere are related to each other; a gradual (steep) temperature decrease with increasing height is observed at ozone-enhanced (reduced) longitude and season. Ozone variability is also large at ozone-enhanced longitudes, but not clear in temperature. In the next subsection, we will investigate the mean ozone concentration and the temperature structure in detail, and after that, we will see the ozone and temperature variabilities in time. Finally, we will investigate the relationship between the variability and the mean state in view of the effect of equatorial wave activity around the tropopause.

3.2. Monthly Mean Ozone and Temperature Structures

[19] To describe the longitudinal and seasonal variations of the mean ozone and temperature structure in the TTL, time-altitude sections of the monthly average ozone mixing ratio in the TUT/TLS at the five stations are shown in Figure 4. Above the tropopause, there is an annual variation of ozone mixing ratios synchronized with the tropopause altitude change, as being highest in northern winter and lowest in summer [e.g., *Seidel et al.*, 2001; *Reid and Gage*, 1996]. This has been recognized as a result of the large-scale extratropical stratospheric pump [e.g., *Holton et al.*, 1995], but recently, *Kerr-Munslow and Norton* [2006] presented a provocative idea that equatorial stationary Rossby wave associated with the tropical convective heating would be important.

[20] Ozone mixing ratios at the tropopause are ~100 ppbv. Below the tropopause, not only large seasonal variations but also the zonal wave structure are observed as described in Figure 2. As shown by *Oltmans et al.* [2001] for the central and eastern Pacific stations and *Thompson et al.* [2003b] for the Atlantic and Africa stations, seasonal variations of ozone in the TUT show minima during northern winter and spring and maxima during summer and autumn at most stations.

[21] At Ascension, the ozone concentration above 14 km is gradually enhanced in the later half of the year, and a contour line with 80 ppbv comes downward around 14 km in October–December. Though the thickness of the ozone TTL is relatively thick throughout the year, the minimum mixing ratio is observed in northern spring with a contour line of 60 ppbv reaching up to ~15 km in April–May. At Nairobi, high ozone mixing ratios >100 ppbv just below the tropopause (>16 km) are observed in northern summer. Another ozone maximum is observed in November above ~13 km, but not so large as in July. On the other hand, at Watukosek, the thickness of the ozone TTL is thin throughout the year. The annual cycle of ozone mixing ratio above ~14 km is clear with being minimum in northern winter when it is a local rainy season. Though the seasonal cycle at Fiji is roughly similar to that of stations at neighboring longitudes (Watukosek and San Cristóbal), a remarkably large ozone enhancement is observed around September. At San Cristóbal, ozone maxima during northern summer-autumn and minima during northern winter-spring are also observed. The annual cycle in the TUT is almost synchronized with the tropopause altitude, as a contour line of 60 ppbv is located at 16 km in November–April and at 14 km in August–September.

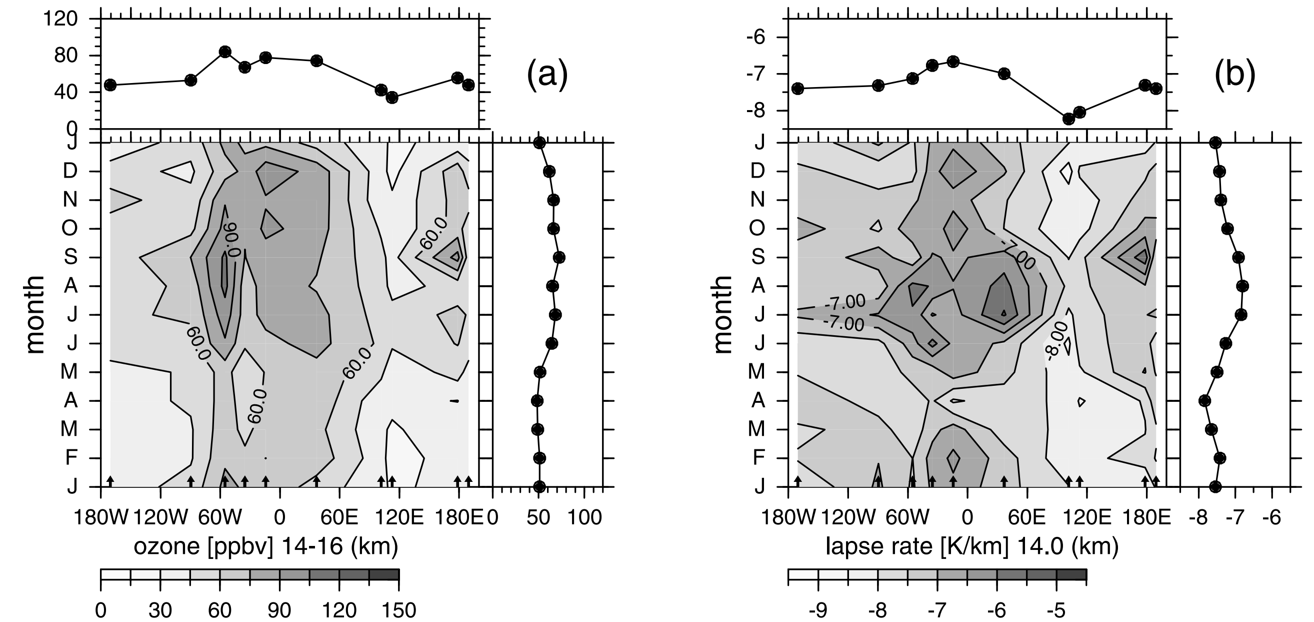

Figure 5. Contour plots of (a) the averaged ozone (ppbv) over 14–16 km and (b) the lapse rate (K km^{-1}) calculated at 14 km. Top and side panels indicate the annual average and the zonal average, respectively.

[22] Figure 5 summarizes the relationship between the background ozone concentration and the lapse rate in longitude and time sections in the TUT. The monthly mean ozone mixing ratio is averaged over the 14- to 16-km height range. The lapse rate is calculated from the 500-m differences centered at 14 km after applying a running mean for 500 m. This altitude is appropriate to investigate the vertical structure of temperature as mentioned above. In Figure 5a we see the zonal wave one structure in the TUT ozone concentration with maxima in the Atlantic and Africa (55°W–37°E) and minima in the western Pacific (102°–113°E) throughout the year. In addition to the zonal maximum around the Greenwich meridian, relatively higher ozone concentration is also observed over the central Pacific (near the date line), though it is smaller than that around the Greenwich meridian. For the annual mean, we observe >70 ppbv at Ascension (14°W), Nairobi (37°E), and Paramaribo (55°W), ~60 ppbv at Fiji, and ~40 ppbv at Watukosek (113°E). The northern summer-autumn maxima and the winter-spring minima are observed at most longitudes, although another ozone enhancement is observed in the Atlantic during late autumn to winter (>90 ppbv in October–December). The ozone enhancement in October–December might be due to pollution [e.g., *Thompson et al.*, 1996] or lightning increases [e.g., *Moxim and Levy*, 2000; *Jenkins et al.*, 2003], but these effects are still not clear yet. We also see a peak of the ozone concentration during summer-autumn at Paramaribo (55°W, 5.8°N). This specific variation may be due to the northern hemispheric origin. The details of the observation at Paramaribo are described by *Peters et al.* [2004]. At Natal (35°W) in March–May, the ozone minimum is observed with <60 ppbv.

[23] Figure 5b shows the longitude-time section of the lapse rate in the TUT, since the ozone concentration and the thermal structure in the TUT seem to be related to each other as mentioned in Figures 2, 3, and 4. In general, we see a good agreement between the two in view of the zonal wave structure with longitudinal maxima around the Greenwich meridian and minima around Indonesia and with seasonal maxima during summer-autumn in the Northern Hemisphere at most longitudes. In the central Pacific, remarkably high stability of >–7 K km^{-1} with the ozone enhancement is observed, but that is restricted to northern autumn.

[24] We have investigated the relationship between the ozone concentration and the lapse rate characterized by the zonal wave one structure and found a general agreement between the two. The east-west contrast of the temperature structure in the TUT could be produced by several factors associated with a tropical heat source. Suppose the equatorial atmospheric response to the heat source [*Highwood and Hoskins*, 1998], there appear a cold anomaly around the tropopause and a warm anomaly in the middle and upper troposphere around the convective region [*Randel et al.*, 2003]. Over the nonconvective region, there appears relatively warm anomaly around the tropopause. Cirrus clouds associated with the cold temperature response may intensify the east-west contrast of the temperature structure [*Norton*, 2001]. As a result, there exist relatively unstable lapse rates centered around 14 km in the convective region and vice versa in the nonconvective region as shown in Figure 5b. Low-ozone anomaly in the TUT can be expected over convectively active longitudes in the tropics where the TUT/TLS vertical transport process is supposed to occur [e.g., *Hatsushika and Yamazaki*, 2003; *Fueglistaler et al.*, 2004] associated with the Rossby and Kelvin wave responses (the Matsuno-Gill response) to the tropical heat source, in which low-ozone air mass in the MBL origin can be transported to the TUT/TLS over the unstable longitude. Thus the longitudinal variations of the temperature structure and ozone concentration can be explained qualitatively, though we may need to consider both radiative and chem-

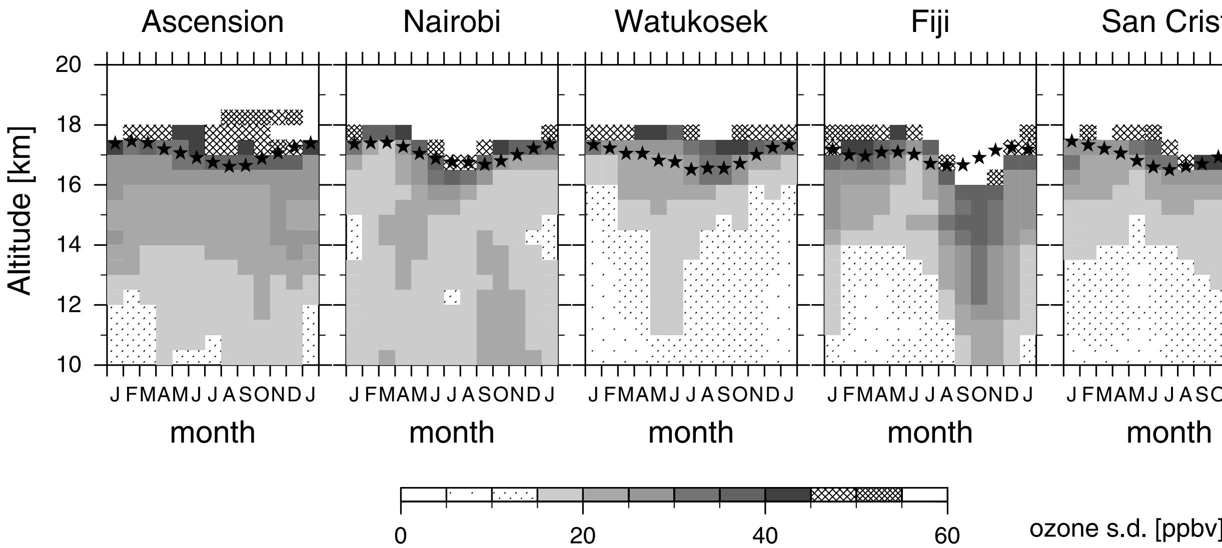

Figure 6. As Figure 4, but for the ozone standard deviation (ppbv) for 10–20 km. Stars indicate locations of the tropopause.

ical effects in the TTL additionally [*Folkins et al.*, 2002; *Folkins and Martin*, 2005]. Moreover, the wave responses can be related with the residual upwelling motions in the TTL as described above [*Kerr-Munslow and Norton*, 2006].

[25] The stability in the TUT can also control the level of deep convection reach; that is, low stability allows penetrative overshooting of convective clouds to higher level [*Reid and Gage*, 1996]. This is consistent with the ozone variation and the temperature structure in both the longitudinal and seasonal sections as shown in Figures 5a and 5b qualitatively.

3.3. Variabilities

[26] In this subsection, we investigate the seasonal variation of ozone variabilities at the five stations as shown in Figure 2. Figure 6 shows time-altitude sections for the standard deviation (SD) of the ozone mixing ratios. We subtracted a monthly mean value from each profile, and binned the data into 3 months and 500-m vertical extent for each SD calculation. Compared with Figure 4 for the monthly mean ozone, relatively large ozone SDs (>15 ppbv) are evident at ozone-enhanced longitudes (Ascension and Nairobi) in the whole upper troposphere. At Ascension in November–January, large ozone SDs are observed above 14 km (~25 ppbv) with enhanced ozone concentrations. Large ozone SDs are also observed at Nairobi in northern summer just below the tropopause (above ~15 km). The ozone SD maxima around 12–15 km during northern spring and those below 14 km during northern autumn are clearly separated from the stratospheric ones, though relatively higher ozone mixing ratios (see 80-ppbv contour line in Figure 4) are observed in November–December. On the other hand, at Watukosek, the ozone variability is small below 15 km (<20 ppbv), but there is a small enhancement of the ozone SD in April–August during the local dry season. At Fiji, large ozone SDs (>25 ppbv) can be seen in northern autumn-winter. At San Cristóbal, ozone SDs in the TUT are slightly high in September–October with enhanced monthly mean ozone concentrations. As already pointed out by *Oltmans et al.* [2001] the ozone variation is larger in the central Pacific than in the eastern Pacific with similar seasonal variations for each other.

[27] As to the temperature SD statistics (not shown), the longitudinal and seasonal variations are not so clear as those for ozone; larger variabilities are observed at specific longitudes and seasons. The details will be discussed in Figure 7b in association with wave activity.

[28] We next present those variabilities in the longitude-time sections in the TUT. Figure 7a shows the longitudinal and seasonal variation of the ozone SDs. For this statistics, we first calculated the ozone SD in the same bin as in Figure 6 and averaged the SDs over the 14- to 16-km range. Generally, the ozone variability is large around the Greenwich meridian (over the Atlantic and Africa; 55°W–37°E) and around the date line (in the central Pacific). This zonal structure looks qualitatively similar to the ozone and lapse rate distributions (Figure 5). The annual average of ozone SD is ~23 ppbv at Ascension and ~26 ppbv at Fiji, in contrast to ~16 ppbv at Watukosek. However, the seasonal variation of ozone SD is not always consistent with the other two variations in Figure 5; the zonal mean ozone SD (in the right panel of Figure 7a) does not show clear seasonality. Moreover, remarkably high ozone variability (>24 ppbv) is observed around the date line during northern autumn-winter, in addition to that in the Atlantic (at Natal and Ascension; >24 ppbv) in northern winter. These maxima are not clearly seen in Figure 5 of the ozone mixing ratio and the lapse rate.

[29] The zonal wave one structure which can be seen in the ozone concentration, the lapse rate, and the ozone variability (Figures 5a and 5b and 7a, respectively) is not clear in the longitude-time section of the temperature SD (Figure 7b). However, remarkably larger temperature variabilities are observed at Nairobi (37°E) during northern summer and at Fiji (178°E) during northern autumn. Another temperature SD maximum is observed at Paramaribo (55°W) in summer.

[30] As the space-time variations of ozone and temperature variabilities investigated in this subsection could be

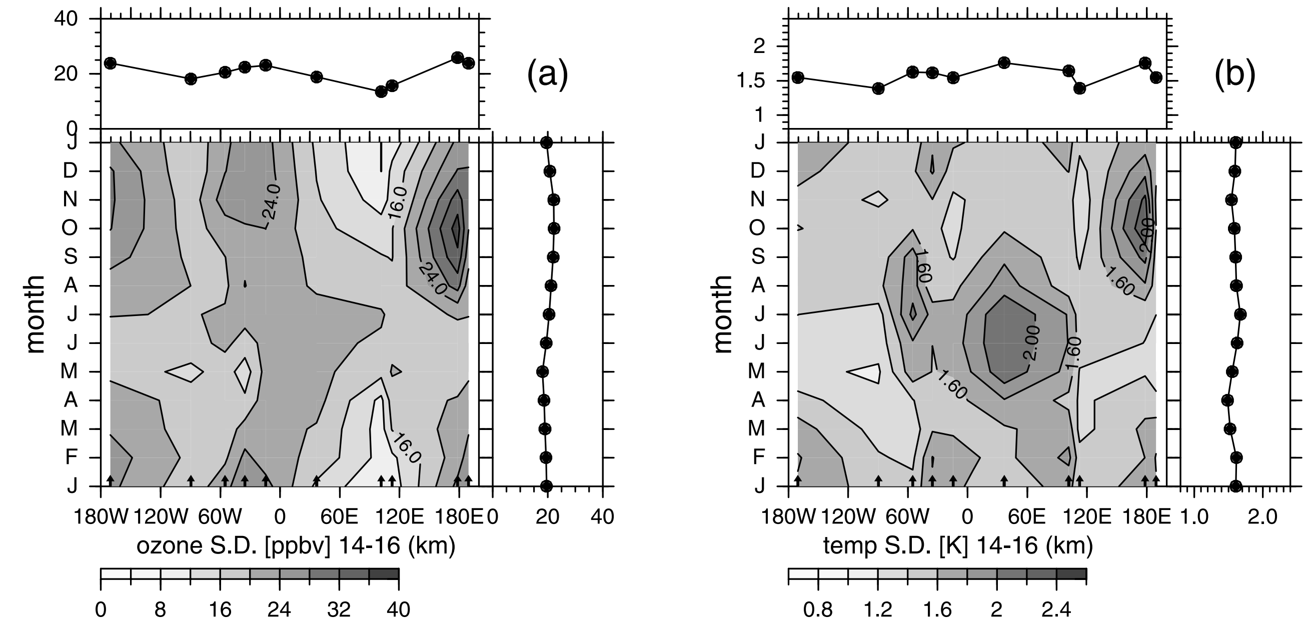

Figure 7. As Figure 5, but for contour plots of (a) the ozone standard deviation (ppbv) and (b) the temperature standard deviation (K) averaged over 14–16 km.

associated with equatorial wave activity, we will next consider the cause of the variabilities. Then we will discuss about possible relations to the equatorial wave activities and the relationship between the variabilities and the means.

3.4. Wave Activities

[31] To consider the relationship between the variability and wave activity, first, we investigate the phase relationship between ozone concentration and temperature around the tropopause. The relation between the two should be coherent in association with large-scale gravity waves or Kelvin waves; if we assume conservation of ozone mixing ratio and potential temperature, both of which have a positive gradient in height [e.g., *Randel*, 1990; *Ogino et al.*, 1997]. Figure 8 shows the longitude-altitude section of correlation coefficients between the ozone mixing ratio and the temperature for JJA. The correlation is good above the tropopause at almost all stations, but it is gradually diminished below the tropopause, though it is still good at some stations.

[32] To summarize the relation between the ozone and temperature variations in the TUT, Figure 9 shows the longitude-time section of the correlation coefficients averaged over the 14- to 16-km height range. Unlike in Figures 5a and 5b, the zonal wave one structure is not clear, but it is rather similar to that of the temperature SD in Figure 7b. Remarkably good correlations are observed at Fiji during northern autumn and at Nairobi during summer, when and where the temperature SDs are generally large. The extent of the ozone SD variation is mostly proportional to the monthly mean ozone value as seen in Figure 2 and in Figures 5a and 5b, but the correlation between the ozone and temperature variations is not always good in the ozone-enhanced longitude and season. Comparing the correlation at Nairobi with that at Ascension in JJA, it is good only at Nairobi, although the ozone variations at both stations are large.

[33] In the tropics, there are the following two kinds of typical large-scale waves: One is the equatorial Kelvin wave propagating eastward with an eastward tilt of the temperature structure, and the other is the equatorial Rossby wave propagating westward. We further examine vertical structures of the temperature and ozone concentration which may be associated with these large-scale waves. The results are

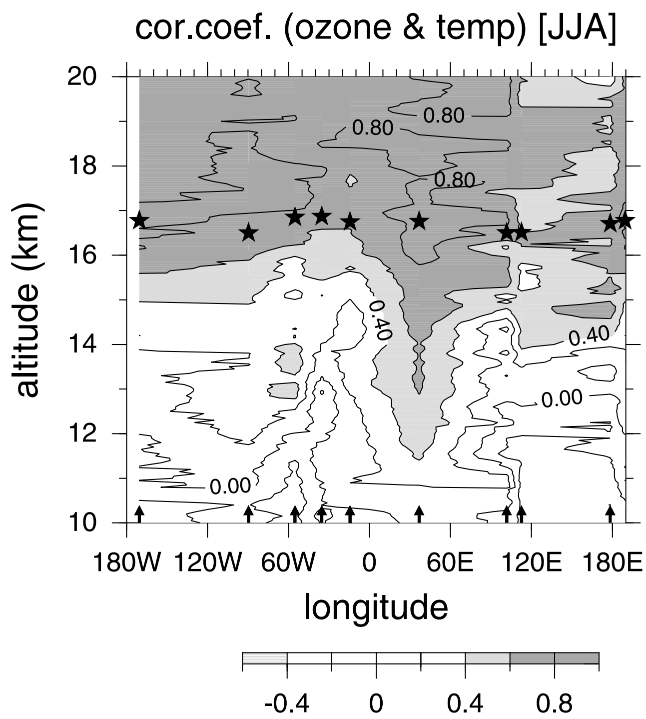

Figure 8. Longitude-altitude section of the correlation coefficient between the ozone mixing ratio and the temperature in JJA.

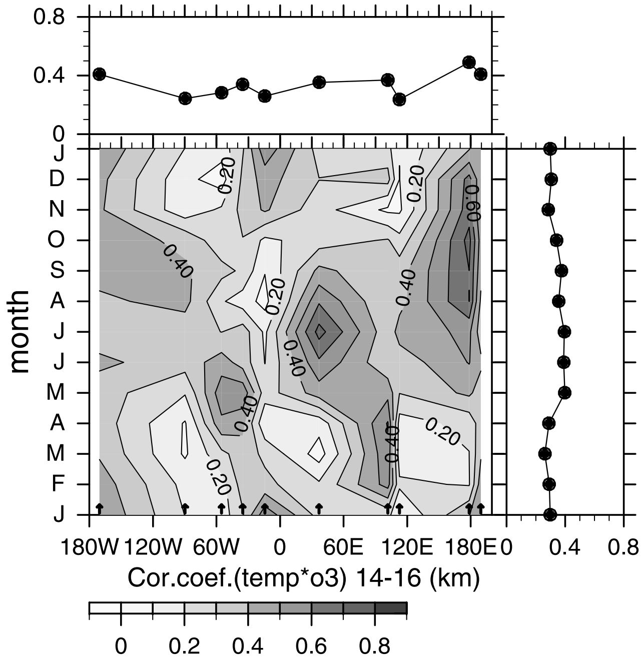

Figure 9. As Figure 5, but for a contour plot of the correlation coefficient between temperature and ozone averaged over 14–16 km.

shown on the bases of a composite analysis for the temperature and ozone concentration as shown in Figure 10. We selected the warmest and coldest profiles for 20% each at three levels of 12, 14, and 16 km and averaged the profiles. Then, we subtracted the average cold profile from the average warm profile and divided by two at each level. For the ozone composite, we use ozone profiles corresponding to the warmest and coldest temperature profiles at the three levels. Here we focus on a case at Nairobi in JJA (Figure 10a) and that at Fiji in September–November (SON) (Figure 10b), because these are two typical cases of a good correlation and large temperature variabilities in the TTL.

[34] In both figures for the temperature composite (solid line in Figure 10), the warmest points are located at the altitude indicated by the horizontal bar. For example, in the rightmost profile in which we used the warmest and coldest profiles at 16 km, the warmest point is found at 16 km by the definition. At Nairobi during northern summer, a wavy shape of the temperature profile with cold and warm maxima is clearly seen. For the ozone distribution at Nairobi during summer, the low-ozone anomalies tilt along with the cold temperature anomalies above 12 km and the high-ozone anomalies correspond to the warm temperature anomalies elsewhere, though the amplitude of ozone increases with altitude associated with background ozone concentration. This type of the structure can be caused by the equatorial Kelvin wave which propagates eastward with an eastward tilt of the temperature anomaly. At Fiji during northern autumn to spring, a good correlation between ozone and temperature variabilities is observed above ~14 km with some ozone-enhanced profiles as seen in Figure 2. However, the temperature structure is somewhat standing with a node around 13–14 km and the ozone structure is rather deep in the TUT, which is quite different from Nairobi in JJA. See right and left profiles at Fiji in Figure 10. A similar structure is also observed at American Samoa and at Tahiti in the central Pacific (not shown). These variations might be associated with a kind of the equatorial Rossby wave, in which the vertical structure is equivalent barotropic [*Kiladis and Wheeler*, 1995; *Wheeler et al.*, 2000]. This type of the equatorial wave is often observed in the central Pacific during northern autumn-spring [*Kiladis and Wheeler*, 1995], where and when we observed high ozone variabilities. Note that the extratropical Rossby wave, which propagates to equatorward under the westerly wind condition, is also active in the central Pacific during northern autumn-spring in the tropics [*Waugh and Polvani*, 2000]. In fact, large negative potential vorticity is sometimes observed at Fiji during northern autumn with an ozone enhancement in the TUT (not shown). However, the distinction between the equatorial Rossby wave activity and the extratropical Rossby wave intrusion may be difficult, because the origin of extratropical Rossby wave may be the extratropical Rossby wave intrusion [*Kiladis*, 1998].

[35] From the results of the temperature SD and the phase relationship between the ozone and temperature variabilities, we can conclude that the zonal wave one structure of ozone may not be understood owing to the wave activities such as the equatorial waves, although large temperature variabilities with a good correlation between the two are observed at some specific times and stations.

4. Summary

[36] Ozone variations with seasonal and intraseasonal timescales in the tropical tropopause layer (TTL) have been investigated using a 5-year tropical ozonesonde data set from the SHADOZ (Southern Hemisphere Additional Ozonesondes) archive by paying particular attention to the longitudinal structure. There is a zonal wave number one structure of ozone in the tropical upper troposphere (TUT)

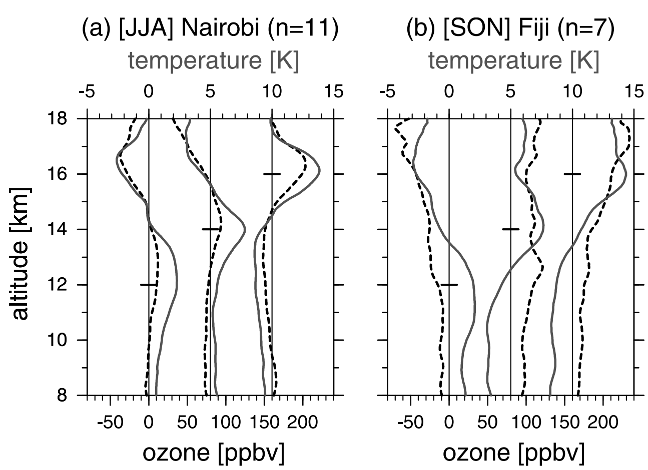

Figure 10. Composited temperature (solid line; K) and ozone (dashed line; ppbv) profiles (a) at Nairobi in JJA and (b) at Fiji in SON. See text for details of the composite method. Individual temperature (ozone) profiles are shifted every 5 K (80 ppbv) from left to right. We use 22 profiles at Nairobi and 14 profiles at Fiji for 40% (20% for each side).

with maxima around the Atlantic and Africa and minima around the western Pacific throughout the year.

[37] We have investigated the ozone distribution and the vertical temperature structure (lapse rate) in the TUT and found that the relationship between the two is good in both longitudinal and seasonal sections. Stable lapse rates are observed around the Atlantic and Africa in longitudinal section and during northern summer to autumn in seasonal section at most stations, while unstable lapse rates are observed in Indonesia and during northern winter-spring. The east-west contrast in the temperature structure and the ozone variation can be explained qualitatively by the large-scale response to the convective heat source. There exists a cold (warm) anomaly around the tropopause and a warm (cold) anomaly below ∼14 km over the convective (nonconvective) region, resulting in unstable (stable) thermal structure over the convective (nonconvective) region. Cirrus clouds associated with the cold temperature may intensify the east-west contrast of the temperature structure [*Norton*, 2001], and at the same time, low ozone air mass in the marine boundary layer origin is transported to the TUT/TLS over the unstable longitude associated with convective activity. Moreover, the atmospheric response can be related with the residual upwelling motion in the TTL [*Kerr-Munslow and Norton*, 2006].

[38] Ozone variability is also large at the ozone-enhanced longitudes (around the Greenwich meridian) and seasons (northern summer-autumn). The variabilities could be caused by wave activities around the tropopause, but the zonal wave structure is not clear in the temperature variability and in the correlation between ozone and temperature. This suggests that the longitudinal variation of the wave activities might not be an important factor in the zonal wave structure of ozone. In the specific longitude and season such as during northern autumn-winter in the central Pacific and during summer in Africa, remarkably large ozone variability is observed with large temperature variability and good correlation between ozone and temperature. From the composite analyses, the large ozone variabilities seem to be associated with the equatorial wave activities, but the relationship between the wave activity and the background ozone is not clear in other longitudes and seasons.

[39] Since we have been interested in rather shallow structures of ozone and temperature around the tropopause, we used ozonesonde data at nine stations around the tropics from the SHADOZ archive. It was found that the data would be useful to reveal the large-scale zonal wave one structure of ozone and temperature in the TTL, but there are still several weaknesses of the data coverage. Some stations used in this study are rather located at subtropical latitudes, and those observations may be highly affected by the midlatitude phenomena. In particular, we could say that the central Pacific is still a data sparse area, though there are SHADOZ stations such as Fiji and American Samoa located around 15°–20°S off the equator. Moreover, the data density is still not enough to investigate the temperature and ozone relation as seen at the specific stations and seasons in Figure 10. We need additional observations near the equator particularly in the central Pacific, and a supplemental use of the satellite data would be useful for future studies. On the other hand, latitudinal transport or mixing between the TTL and midlatitude may be one of the possible ozone sources in the TTL, although we could not investigate the effect in this study because of the sparseness of the observations in both longitudinal and latitudinal directions. The future investigations at additional sites through continuous observations may clarify the relationship between the horizontal mixing and the ozone variation in the TTL.

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