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## Ozonesonde observations in the Indonesian maritime continent: a case study on ozone rich layer in the equatorial upper troposphere

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### Abstract

Ozonesonde observation campaigns were conducted over the Indonesian maritime continent in September–October 1998 and in August–September 1999. Three stations were used for each campaign, Watukosek (7.5°S, 112.6°E), Kototabang (0.20°S, 100.3°E), and Pontianak (0.03°N, 109.3°E) for the 1998 campaign, and Watukosek, Kototabang, and Darwin (12.25°S, 130.55°E) for the 1999 campaign. Both periods were basically characterized as the La Niña period, and the tropospheric ozone concentrations showed normal values. Temporal variation and horizontal distribution of an ozone layered structure with a 1–1.5-km thickness were obtained just below the tropopause at the two equatorial stations during the 1998 campaign. Meteorological data analyses including the reverse domain filling technique suggested that the most plausible explanation for the layer is the quasi-horizontal, thin intrusion from the northern midlatitude lower stratosphere associated with a breaking Rossby wave and large-scale flow pattern.

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### 1. Introduction

Ozonesonde soundings and total ozone measurement with the Brewer spectrophotometer have been conducted at Watukosek (7.5°S, 112.6°E), Indonesia since

1992 and since 1993, respectively (Komala et al., 1996; Fujiwara et al., 2000). These ozone observations have revealed the seasonal and interannual variabilities of tropospheric ozone in this region. During the local wet season, December to March, the ozone mixing ratios are nearly constant at 25 ppbv throughout the troposphere; and during the local dry season, August to November, the concentrations are enhanced in the planetary boundary layer (PBL). In the September–October periods of 1994 and of 1997, El Niño and Indian Ocean Dipole (IOD) events were in phase (Saji et al., 1999),

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causing severe droughts and widespread, natural and agricultural biomass burnings in Indonesia. (IOD is an independent atmosphere–ocean coupling oscillation in the tropical Indian Ocean with a similar periodicity to the El Niño–La Niña in the Pacific Ocean.) These burnings resulted in pronounced enhancements of the ozone mixing ratio in the middle troposphere, integrated tropospheric ozone, and total ozone at Watukosek (Fujiwara et al., 1998b, 1999). Satellite data analyses have also characterized these events (Chandra et al., 1998; Kita et al., 2000; Thompson et al., 2001). The influence of the Indonesian burnings on tropospheric ozone was also reported from Malaysia (Yonemura et al., 2002a) and from Singapore (Yonemura et al., 2002b). The Watukosek ozonesonde observations have also revealed a new mechanism of the stratosphere–troposphere exchange (STE) associated with the equatorial Kelvin wave (Fujiwara et al., 1998a; see also Fujiwara et al., 2001; Fujiwara and Takahashi, 2001).

The Biomass Burning and Lightning Experiment (BIBLE) phases A (September–October 1998) and B (August–September 1999) were conducted for estimating the photochemical impact of the Indonesian maritime continent by using an aircraft equipped with the instruments for measuring ozone and its precursors (Kondo et al., 2001, 2002). During these campaigns, we also made quasi-simultaneous ozonesonde observations at three stations in the maritime continent for the first time, coordinating as to obtain vertical and horizontal ozone field when the aircraft was operated (Fig. 1 and Table 1). The ozonesonde results provided not only the general ozone field during the two periods but also the temporal variation of some layering structures.

Recently, either horizontally or vertically fine atmospheric structures in the troposphere have drawn much

attention (e.g., Appenzeller et al., 1996; Newell et al., 1999; Stoller et al., 1999; Thouret et al., 2000, 2001; Shiotani et al., 2002). The omnipresence of fine-scale filaments and layers is a basic nature of the atmosphere, characterizing the mixing time scale between two air masses such as the stratospheric and tropospheric air masses and the tropical and extratropical air masses. Thus, these fine-scale structures should be taken into account to assess the budget of minor constituents. Some of them are a part of the well-known atmospheric waves or eddies, and some may be independent phenomena. But, especially in the tropics, many of them have not been fully understood yet in terms of the atmospheric dynamics. Therefore, it is still very important to measure the temporal variation of fine-scale structures and to characterize the specific process for producing each of them.

In this paper, we first describe the ozonesonde observations (Section 2) and characterize the two campaign periods in terms of the ozone concentration over the maritime continent (Section 3.1). Then we focus on a case of ozone layered structures observed in the equatorial upper troposphere (Section 3.2). Section 4 summarizes the findings.

## 2. Observations

Table 1 summarizes the ozonesonde observations conducted in September–October 1998 and in August–September 1999. Following Fujiwara et al. (1999, 2000) and Kita et al. (2000), tropospheric ozone in Dobson unit ( $1 \text{ DU} \equiv 2.687 \times 10^{16}$  molecules of ozone  $\text{cm}^{-2}$ ) is defined as the ozone amount integrated from the surface to 15 km in this paper. The ozone amount between

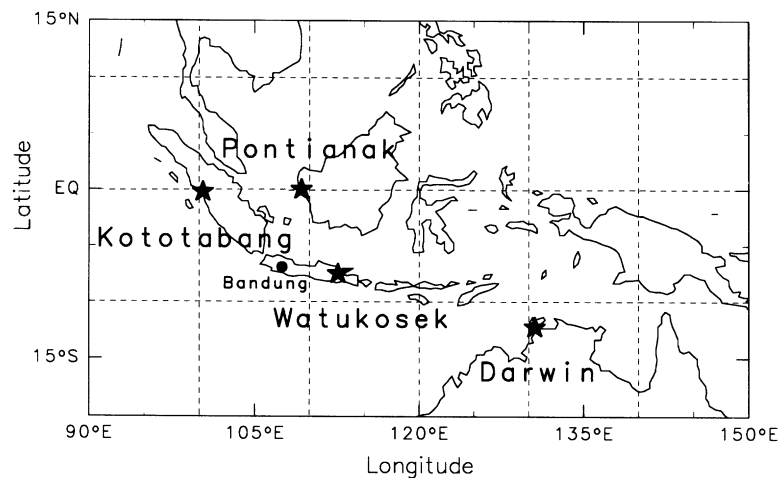


Fig. 1. Map of the Indonesian maritime continent. The locations of the ozonesonde stations are indicated by ★. Bandung (●) is the aircraft base for the BIBLE-A campaign.

Table 1  
Summary of ozonesonde observations

Station (Ozonesonde type)	Date	Tropospheric ozone (DU)
<i>September–October 1998</i>		
Watukosek (7.5°S, 112.6°E; 50 m) MEISEI RSII-KC79D	Sep. 23, 29, 30, Oct. 6, 7	19.3±8.1
Kototabang (0.20°S, 100.3°E; 865 m) EN-SCI ECC, buffered 0.5% KI	Sep. 29, 30, Oct. 2, 6, 7	12.0±1.0
Pontianak (0.03°N, 109.3°E; 20 m) Science Pump ECC, buffered 1% KI	Sep. 16, 23, 27, 29, 30, Oct. 2, 6, 7, 9	17.4±3.0
<i>August–September 1999</i>		
Watukosek EN-SCI ECC, unbuffered 2% KI	Aug. 25, 31, Sep. 4, 7, 10, 13	22.5±2.9
Kototabang EN-SCI ECC, unbuffered 2% KI	Aug. 31, Sep. 2, 4, 7, 10, 13	18.9±3.3
Darwin (12.25°S, 130.55°E; 30 m) EN-SCI ECC, unbuffered 2% KI	Sep. 2–4, Sep. 6, 7, 10	30.4±2.5

15 km and the tropopause (typically located around 16 km) is negligibly small (<1 DU). Different types of ozonesonde and potassium iodide (KI) solution were used. However, all the electrochemical concentration cell (ECC) ozonesondes used here can be regarded as identical for the present purpose, with an accuracy of 5–10% in the troposphere (Komhyr et al., 1995; Oltmans et al., 2001). The difference in ozone concentrations at different stations is mainly due to the geographical variability rather than the difference in sonde type. At Watukosek we used electrochemical carbon–iodine ozonesondes, MEISEI RSII-KC79D (Kobayashi and Toyama, 1966), until July 1999, and then switched to the ECC ozonesondes. In the troposphere the quality of the MEISEI ozonesonde is basically similar to the ECC ozonesonde or may be somewhat worse. See Thompson et al. (2002) for comparison of the ozonesonde performance in the tropics. As for the relative humidity (RH) measurement, we used Vaisala RS80 A-Humicap sensors for all the ECC measurements. The measured RH is always with respect to liquid water, and the measurement is valid without any corrections down to  $-30^{\circ}\text{C}$  air temperature (up to  $\sim 10$  km in the tropics) (e.g., Miloshevich et al., 2001). The MEISEI system has no humidity sensor.

For the BIBLE-A campaign, in September–October 1998, the aircraft base was at Bandung (6.9°S, 107.5°E). The three ozonesonde stations were chosen to surround the aircraft base and some flight tracks and provided the information of horizontal and vertical ozone distribution basically on the days of the aircraft measurement. For the BIBLE-B campaign, in August–September 1999, the aircraft base was at Darwin. Ozonesonde soundings at Darwin were conducted basically on the days of the

aircraft measurement, and the two Indonesian stations, which were located upstream for Darwin in the middle to upper troposphere, provided the general information on the ozone distribution over the western maritime continent.

### 3. Results and discussion

#### 3.1. Ozone distribution in the maritime continent

Fig. 2 shows the tropospheric ozone amounts integrated from the surface to 15 km at Watukosek for 1998 and 1999 and at the special stations during the campaign periods. The very strong El Niño event from 1997 to early 1998 (e.g., Fig. 6 of Fujiwara et al., 2000), being concurrent with an IOD event, terminated around May 1998 (recognized by the Southern Oscillation Index from Climate Prediction Center, US National Oceanic and Atmospheric Administration; see <http://www.cpc.noaa.gov/data/indices/>). A La Niña event then started around June 1998 and lasted until the middle of 1999. Therefore, the period from September to October 1998 corresponds to a La Niña period when the maritime continent is expected to be relatively wet and poor in ozone, and the period from August to September 1999 corresponds to a normal period or a weak La Niña period. In early 1998, the tropospheric ozone at Watukosek was still recovering from the pronounced enhancement ( $\sim 55$  DU) due to the El Niño-IOD-induced extensive biomass burning in September–October 1997 (see Fujiwara et al., 2000). After April 1998, the tropospheric ozone was about 20 DU at Watukosek, a normal value at this station (Fujiwara et al., 2000), with an increase up to about 30 DU around the two campaign periods,

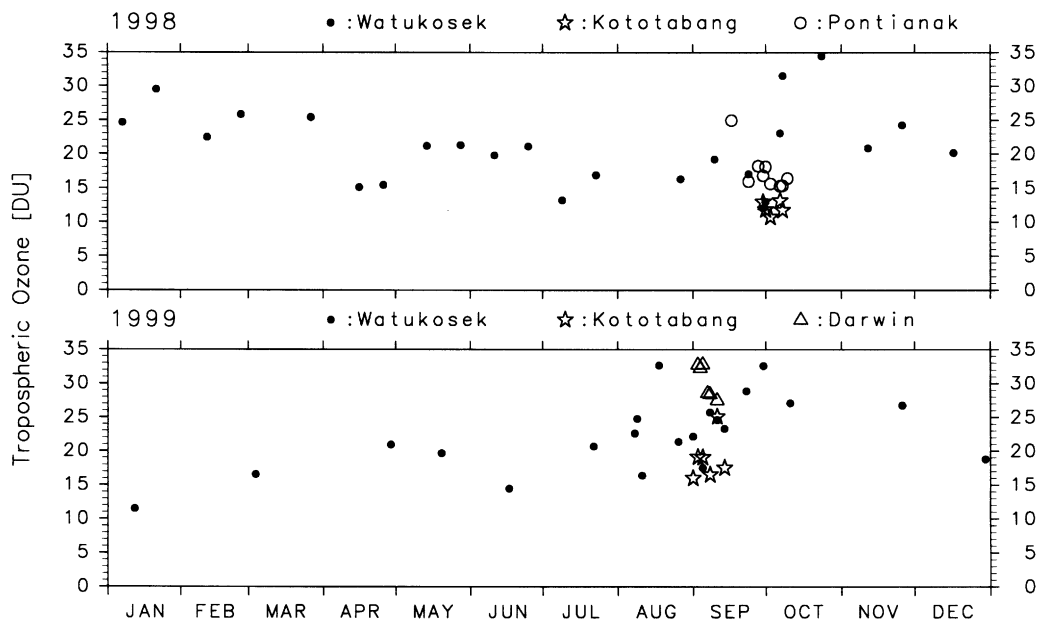


Fig. 2. Variations of tropospheric ozone integrated from the surface to 15 km (top) in 1998 and (bottom) in 1999. Dots are for the Watukosek observation. Open stars are for Kototabang. Open circles in 1998 are for Pontianak and open triangles in 1999 are for Darwin.

August–October 1998 and 1999, when it is the dry season in the southern hemispheric part of the maritime continent.

Table 1 also summarizes the average values of the tropospheric ozone with their standard deviation during the campaign periods. Although the variability at Watukosek in 1998 is large, the average tropospheric ozone in the 1999 campaign exceeds that in the 1998 campaign both at Watukosek ( $\sim 20\%$ ) and at Kototabang ( $\sim 60\%$ ). The sounding results in the 1999 campaign suggest that the concentration tends to increase from the equator to the south. This is consistent with the facts that the influence of the southern subtropical air, with higher ozone concentrations, is greater at the off-equatorial stations than at the equatorial stations and that the equatorial stations during the campaign periods were in the rainy season (shown later).

Fig. 3 shows the average profiles of ozone, temperature, and RH in the troposphere. The Watukosek plots are for the August–November average, and the 1998 Watukosek plot also contains the 1993–1996 average profile taken from Fujiwara et al. (2000). The RH profiles clearly show a complicated wet/dry season distribution within the maritime continent (see also Hamada et al., 2002). The equatorial stations (Kototabang and Pontianak) were in the wet season during the two campaign periods, but the off-equatorial stations (Watukosek and Darwin) were in the dry season. The middle troposphere over Darwin was especially dry, with  $<10\%$  RH. All the average ozone profiles have

nearly constant concentrations in the troposphere except the PBL region, although many of the soundings captured layered structures with a thickness of 0.5–3 km especially at Darwin and Watukosek during the 1999 campaign (not shown). The Watukosek profiles in both years are comparable to the dry-season profile at this station. The Kototabang and Pontianak profiles, which have concentrations of 20–30 ppbv throughout the troposphere, resemble the wet-season profile at Watukosek except the PBL region (Fujiwara et al., 2000). The Darwin profile has a concentration of 40 ppbv above 5 km and a slightly ozone-enhanced, dry layer at 2–4 km, just above the PBL. There is a general tendency that the wet (dry) station has lower (higher) ozone concentrations. Note that the standard deviation of the 1998 Watukosek ozone profile (taken by the MEISEI ozonesondes) was two to three times larger than the other observations (by the ECC ozonesondes), especially above 10 km. The investigation of the individual profiles suggests that this may be attributed partly to the difference in the observing system.

### 3.2. Layered structures in the equatorial 15-km region in September–October 1998

During the 1998 campaign, an interesting set of ozone layered structures was obtained just below the tropopause at Pontianak and Kototabang from 29 September to 2 October (Kototabang) and to 6 October (Pontianak) (Fig. 4). The layer appeared on 29 September at the

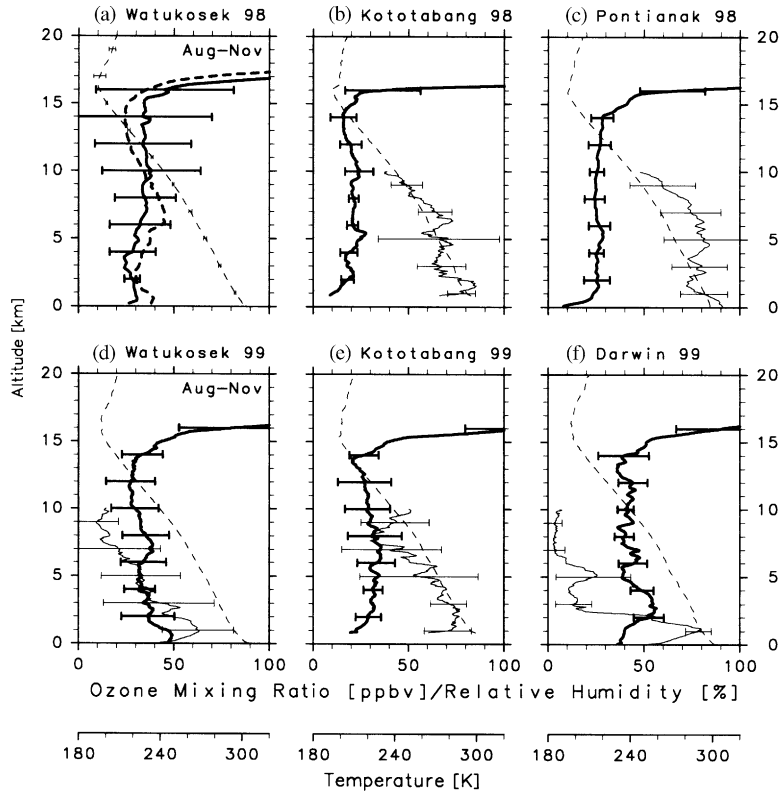


Fig. 3. Average profiles of ozone mixing ratio (thick curves), temperature (thin dotted curves), and RH (thin curves) for: (a) Watukosek in August–November 1998; (b) Kototabang in September–October, 1998; (c) Pontianak in September–October, 1998; (d) Watukosek in August–November, 1999; (e) Kototabang in August–September, 1999; and (f) Darwin in August–September, 1999. The average ozone profile at Watukosek in August–November 1993–1996 is indicated by a thick dotted curve in (a). The horizontal bars represent the standard deviation.

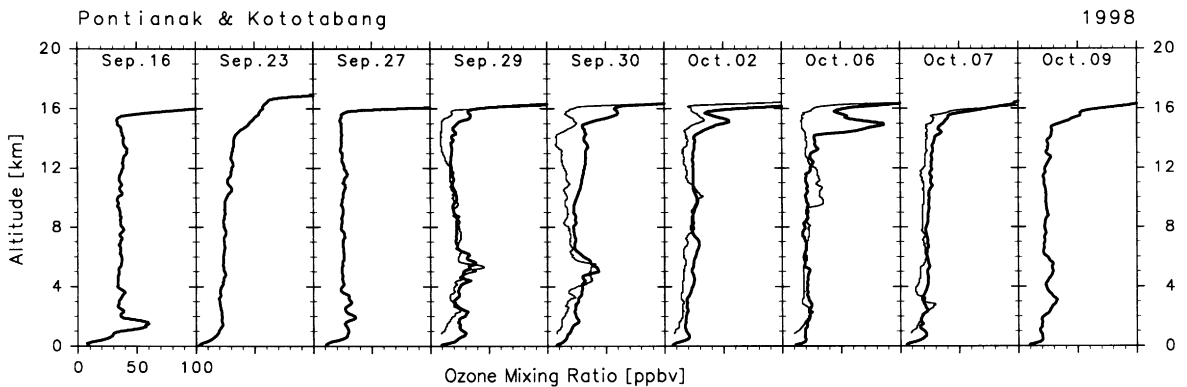


Fig. 4. Tropospheric ozone profiles at Pontianak (thick curves) and at Kototabang (thin curves) in September–October 1998.

both stations. Its peak concentration increased gradually but its peak altitude decreased slightly, especially at Pontianak, and the layer vanished by 6 October at Kototabang and rather abruptly on 7 October at Pontianak. The peak concentration of the layer was always higher at Pontianak than at Kototabang during

the period. The maximum layer thickness was 1.5 km. On 6 October, a maximum concentration of 86 ppbv was observed at Pontianak at 14.9 km. This mixing-ratio value was much larger than those lower in the troposphere during this period, suggesting that the origin of the air mass was in the stratosphere or at a rather

distant, highly polluted surface region. As for the latter, however, we need extremely strong emission of ozone precursor gases similar to the 1997 Indonesian burning episode (Fujiwara et al., 1999) to obtain a concentration of 86 ppbv in the upper troposphere. A major contribution of lightning-produced  $\text{NO}_x$  may also be excluded for this case because the ozone production rate including the lightning process was estimated as  $1.4 \text{ ppbv d}^{-1}$  at most in the upper troposphere over the maritime continent (Pickering et al., 1993). The apparent life time of the layer, about a week, suggests that the phenomenon may have been associated with synoptic- or planetary-scale meteorological disturbances.

The following three processes are investigated as possible causes.

- Transport from the tropical stratosphere associated with large-scale equatorial gravity waves including equatorial Kelvin waves (Fujiwara et al., 1998a, 2001; Fujiwara and Takahashi, 2001)
- Isentropic transport from the midlatitude lower stratosphere to the tropical upper troposphere associated with breaking Rossby waves (e.g., Chen, 1995; Postel and Hitchman, 1999; Waugh and Polvani, 2000)
- Vertical transport from a rather distant, polluted surface region associated with deep convections (e.g., Thompson et al., 1997)

Radiosonde sounding data with high vertical resolution are useful for investigating process (a). An intensive radiosonde observation campaign, two launches per day, was conducted at Pontianak from 26 August to 21 October. Fig. 5 shows the potential temperature variation in the 12–20-km region. Downward transports associated with large-scale gravity waves around the tropopause can be seen from 9 to 13 September and from 8 to 17 October. For each case, a significant tropopause jump of  $\sim 2 \text{ km}$  is followed by a descending motion visualized by the isentropes' descent just below the tropopause. However, the ozone layered structure

appeared from 29 September to 6 October, the period when the gravity wave activity was low around the tropopause (i.e., the tropopause-height variations were within 1 km without jumps), so that the process (a) cannot explain the layer.

Trajectory analyses with global meteorological analysis data may be useful for investigating processes (b) and (c). The isentropic trajectory model used here was developed at Earth Observation Research Center, National Space Development Agency of Japan. The kinematic trajectory model (using the vertical wind data) used here was developed at National Institute of Polar Research, Japan (Yamanouchi et al., 2002). The European Centre for Medium-Range Weather Forecast (ECMWF) global analysis data were used as the input for both calculations. Fig. 6 shows isentropic and kinematic backward trajectories starting from Pontianak at 12–17 km on 6 October when the maximum concentration was observed at this station. The results for Kototabang were basically the same as those for Pontianak. The backward trajectories from the three ozonesonde stations confirmed that the air mass in the upper troposphere over western Indonesia in this period basically came from northeast, that is, from the Philippines Islands and the western Pacific south of Japan. (Wtukosek was sometimes influenced by the southern subtropical air as well.) It took  $\sim 4$  days for the trajectories to move a  $20^\circ$  latitudinal range. However, the vertical motions differ between the isentropic and kinematic trajectories. The isentropic trajectories maintain their altitudes almost the same as those at the calculation starting point, but the kinematic trajectories move gradually downward as moving backward. Some trajectories reach as low as 3 km, near the top of the tropical PBL. This upward air mass motion moving toward the equator might be due to the convective upward motion. The geostationary meteorological satellite (GMS) cloud data show that cloud clusters with a diameter of 1000 km or more were common over the region where the trajectories south of

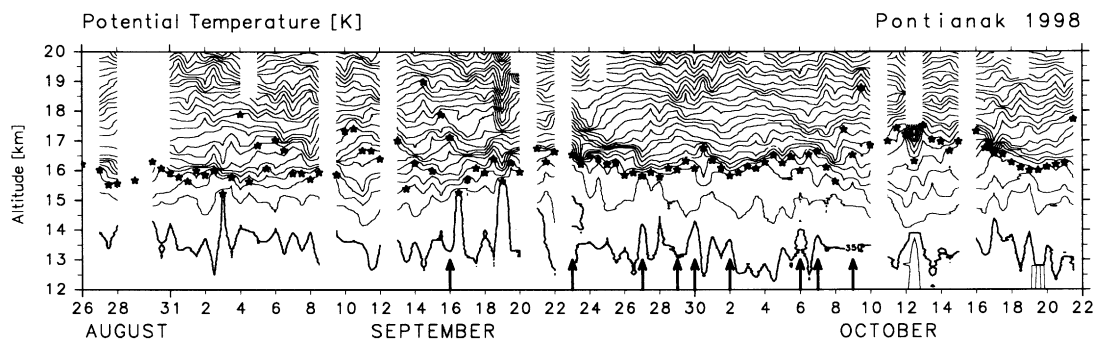


Fig. 5. Time-altitude distributions of potential temperature at Pontianak between 26 August and 21 October, 1998 in the 12–20-km region. The contour interval is 5 K. The thick curves are for 350 K. The location of the tropopause defined by the temperature minimum is indicated by stars. The arrows indicate the ozonesonde soundings at this station.

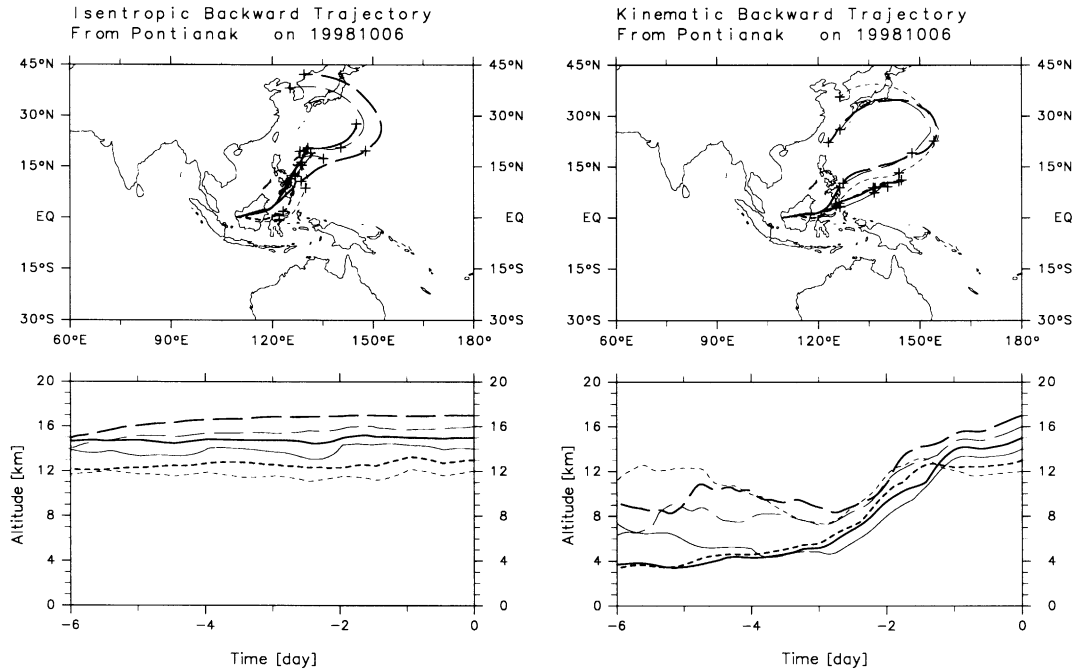


Fig. 6. Results of 6-day backward trajectory analyses from Pontianak at 12 km (thin dotted curves), 13 km (thick dotted curves), 14 km (thin curves), 15 km (thick curves), 16 km (thin broken curves), and 17 km (thick broken curves) on 6 October, 1998. Left panels are for the isentropic calculation, and right panels are for the kinematic calculation. Top panels indicate the horizontal motions, and bottom panels indicate the vertical motions. Crosses on the trajectories in the top panels show the 2-day interval.

15°N traced in this period (not shown). The isentropic trajectories suggest that the enhanced ozone observed just below the tropical tropopause may have originated in the northern midlatitude lower stratosphere (process (b)). The kinematic trajectories suggest that the enhanced ozone may have originated near Philippines Islands (process (c)), although, as previously noted, it is rather hard to explain the peak concentration (86 ppbv) of the observed ozone enhancement by this process.

Potential vorticity (PV) analysis is often used to investigate process (b) because high (absolute) PV regions in the extratropical troposphere can be regarded as being affected directly by the stratospheric air intrusion (e.g., Holton et al., 1995). Although this is an equatorial case, the PV analysis may show some evidence of the transport from the midlatitude lower stratosphere. However, the PV distributions calculated directly from the global analysis data sometimes lack the fine-scale structures that in situ measurements do capture. One of the methods that may overcome this problem is the reverse domain filling (RDF) backward trajectory technique (Sutton et al., 1994; Newman and Schoeberl, 1995). The procedure of the RDF technique is as follows. First, points are distributed in the area of interest (the 40°S–40°N, 90°E–150°W area on the 355-K potential temperature surface, in this study) with an

arbitrary spatial resolution ( $1^\circ \times 1^\circ$ , in this study), generally finer than that of the input data ( $2.5^\circ \times 2.5^\circ$  for the ECMWF data). Second, the backward trajectories are calculated from all these points for an arbitrary time period (as far as the PV of the air mass is considered to be preserved). Finally, the PV values are calculated at the end of these trajectories and projected on to the initial area to reconstruct the PV field. In this study, the isentropic backward trajectories were calculated for  $td$  days, where  $td = 1, 2, \dots, 7$ , to derive the PV fields (referred to as the  $td$ -RDF calculation). Fig. 7 shows the PV maps on the 355-K isentropic surface on 2–7 October from the 3-day RDF calculation. The directly calculated PV maps have no remarkable high-PV structure affecting the two equatorial stations (not shown), but the 3-day RDF PV maps show that high PVs extending from the central North Pacific did reach the two stations during the period. Postel and Hitchman (1999) pointed out that the North Pacific is one of the regions where breaking Rossby waves cause rich interaction between the tropical troposphere and extratropical stratosphere (see also a model study by Horinouchi et al., 2000). The disappearance of the layer at the two stations around 6–7 October is also captured by the 3-day RDF calculation. Note that compared with shorter or longer trajectory calculations, the 3-day RDF result was the most consistent with the ozonesonde

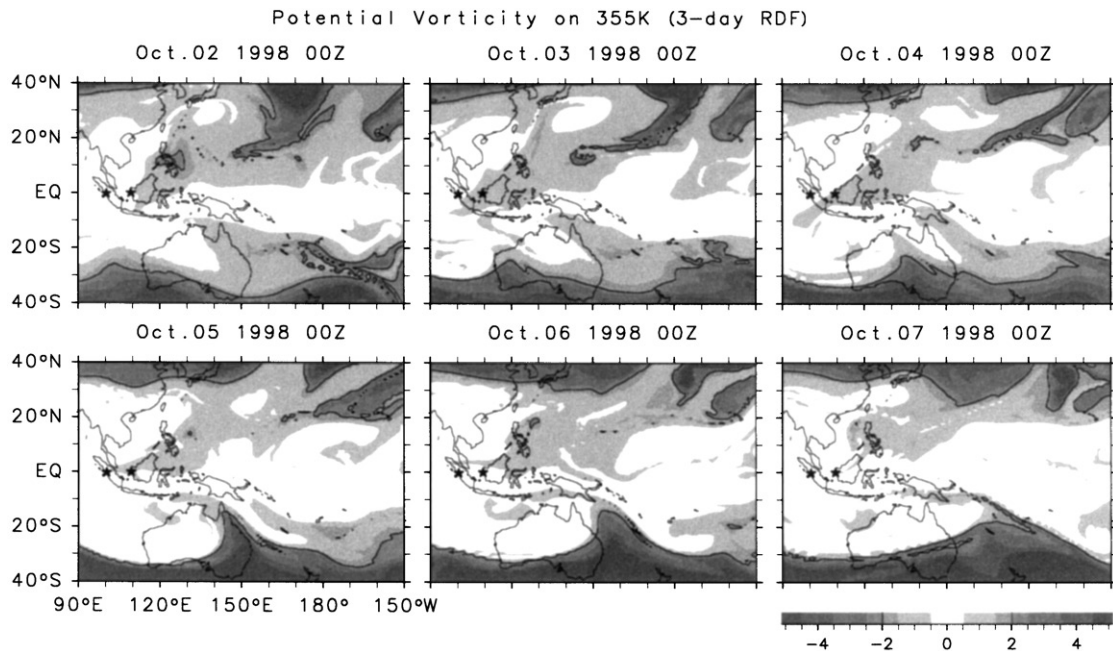


Fig. 7. PV distributions on the 355-K isentropic surface on 2–7 October 1998 by the 3-day RDF calculations (see text for the details). The unit is pvu, where  $1 \text{ pvu} = 10^{-6} \text{ m}^2 \text{ s}^{-1} \text{ K kg}^{-1}$ . The contour lines for  $\pm 2$  pvu are also indicated. Pontianak and Kototabang are indicated by stars.

results in terms of the timing of the high PV passage over the two stations.

In summary, the most plausible explanation for the layered structure observed at Pontianak and Kototabang in September–October 1998 is the transport from the northern midlatitude lower stratosphere associated with the Rossby wave activity over the central North Pacific and with the large-scale flow pattern in the upper troposphere over the subtropical western Pacific and the maritime continent. This is supported by the time scale of the phenomenon, the isentropic trajectory analysis, and the RDF PV analysis. It should be noted that the quasi-horizontal, thin (1–1.5 km) intrusion was observed right at the equator when the activity of large-scale equatorial gravity waves was low around the tropopause. This may mean that the tropical atmosphere is full of layers if convections and convectively generated waves do not exist (cf., Thouret et al., 2001; Shiotani et al., 2002). Although similar observations have already been reported in the tropical Atlantic (e.g., Scott et al., 2001) and in the tropical Indian Ocean (e.g., Zachariasse et al., 2001), this is the first minor-constituent observation around the maritime continent that captured an intrusion event from the midlatitude lower stratosphere to the tropical upper troposphere. It should also be noted that the equatorial 15-km region is within the tropical tropopause/transition layer (TTL) or the tropical substratospheric transition zone in which transport and dehydration processes are thought to be

of crucial importance to determine the characteristics of the tropical lower stratospheric air mass (e.g., Highwood and Hoskins, 1998; Folkins et al., 1999; Haynes and Shepherd, 2001). The present study showed a case of horizontal transport within TTL.

#### 4. Summary

We conducted ozonesonde observations at three stations in the Indonesian maritime continent in September–October 1998 and in August–September 1999. Both campaign periods basically corresponded to a La Niña period, and the average tropospheric ozone concentrations were at a normal level, without significant enhancement as in 1994 and in 1997.

We obtained an ozone layered structure with a 1–1.5-km thickness around 15 km over Pontianak and Kototabang appearing from 29 September to 6 October 1998. Meteorological data analyses including the RDF backward trajectory technique suggested that the most plausible explanation for the layer is the quasi-horizontal, thin intrusion from the northern midlatitude lower stratosphere associated with a breaking Rossby wave and large-scale flow pattern. It should be noted that the layer existed when the gravity wave activity was low around the equatorial tropopause. Active air mass exchange between the midlatitude lower stratosphere and the tropical upper troposphere has been suggested



mostly by the analyses of the global meteorological analysis data (e.g., Chen, 1995; Postel and Hitchman, 1999; Waugh and Polvani, 2000). Field measurements focusing on this process are still very limited in the tropics (e.g., Scott et al., 2001; Zachariasse et al., 2001). The present study suggested a fate of such an air mass originating from the subtropical tropopause region as a thin layered structure extending to the equatorial region. If convection is not active, such a layer can maintain its identity for several days, affecting the budget of minor constituents.

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