Pergamon

Atmospheric Environment Vol. 32, No. 2, pp. 159–168, 1998

1352-2310/98 $17.00 + 0.00

PII: S1352-2310(97)00320-8

ANALYSIS OF THE SEASONAL BEHAVIOR OF TROPOSPHERIC OZONE AT HONG KONG

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(*First received* 8 *November* 1996 *and in final form* 19 *June* 1997. *Published November* 1997)

Abstract—Ozonesondes have been launched at Hong Kong (22.2°N, 114.3°E) since 1993. The results of data analysis of the ozone profiles are presented, with a focus on the seasonal cycle in tropospheric ozone. The results show that the tropospheric ozone column has an obvious maximum in spring and a minimum in summer. The former is a common feature at many locations in the Northern Hemisphere. The summer minimum is attributed to the onset of the summer monsoon when air flow from the Asian continent is replaced by air from the South China Sea or the tropical Pacific. The tropospheric ozone has an important influence on the seasonal cycle of total ozone at Hong Kong. The seasonal cycle of ozone mixing ratio below 2 km is bimodal with ozone peaks in spring and autumn. A frequently observed feature in late autumn and winter is a relative minimum of ozone mixing ratio (as low as 30–40 ppbv) in the upper troposphere (from about 9 to 16 km). Trajectory analysis shows this relative minimum of ozone is associated with air masses coming from the tropical region. It is proposed that the East Asia local Hadley circulation is responsible for this feature. © 1977 Elsevier Science Ltd.

Key word index: Seasonal variation, tropospheric ozone column, vertical ozone profile, relative minimum of ozone mixing ratio, East Asia local Hadley circulation.

1. INTRODUCTION

Ozone is of great interest to atmospheric scientists because it plays an important role in controlling the chemical composition, air quality (WMO, 1994; Tang and Madronich, 1995) and climate evolution of the troposphere. At high concentrations it also has adverse effects on human health, terrestrial plants and outdoor materials. Specifically, changes in tropospheric ozone which reacts with water vapor to form the hydroxyl radical (OH), influence the concentration of many pollutants which are removed from the troposphere by reaction with OH. Ozone can absorb thermal radiation at 9.6 μm and has an effect on the radiative budget of the troposphere (Fishman *et al.*, 1979), especially in the upper troposphere. In the lower troposphere or the atmospheric boundary layer, ozone is known to contribute to crop and tree damage (Skarby and Sellden, 1984) and human health impairment (Lippmann, 1991). The behavior of tropospheric ozone is thus of significant importance.

Tropospheric ozone has been widely studied in other regions (e.g. Logan, 1985; Logan and Kirchhoff, 1986; Volz and Kley, 1988; Oltmans *et al.*, 1989; Oltmans and Levy, 1994), but there are only a limited number of reports about the behavior of tropospheric ozone in the East Asia region. Logan (1985) reported that a summer maximum of tropospheric ozone cannot be found at lower latitudes over Japan (Kagoshima, 32°N) where ozone decreases dramatically in early summer. This was suggested to be related to seasonal changes in atmospheric circulation. Ogawa and Miyata (1985), Tsuruta *et al.* (1989) and Sunwoo *et al.* (1994) reported a similar seasonal variation of surface ozone in Japan, showing a spring maximum and a summer minimum. Seasonal variability of tropospheric residual ozone with highest values in spring and summer over the West Pacific rim region was reported by Sunwoo *et al.* (1992). Akimoto *et al.* (1994) found that 0–2 km layer ozone

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had increased about 2% every year in the past two decades through analysis of ozonesonde data in Japan. Tsutsumi and Makino (1995) used a series of aircraft measurement data obtained from 1987 to 1991 to analyze the vertical distribution of tropospheric ozone over Japan. The results showed that the ozone peaks over Japan occasionally originated from tropopause folding which occurred to the west of Japan. The authors suggested that not only intrusion from the stratosphere but diffusion during transport was also important for the ozone distribution over Japan. Surface ozone data obtained at remote island stations in the East Asian Pacific rim region during the PEM-WEST. A campaign (September–October, 1991) were analyzed by Akimoto *et al.* (1996). Results showed that ozone concentrations at the three sites were characterized by long-range transport from the continent or the ocean.

In the past, most of the ozonesonde stations have been located outside of East Asia. There are four ozonesonde stations in Japan where ozone vertical profiles have been accumulated for more than 30 years. Ozonesonde observations began in Taiwan in July 1992 (Liu *et al.*, 1995). Mainland China has only a few ozonesonde observations before October 1995, and about 50 ozonesondes were launched in Xining (36.2°N, 100.5°E) of Qinghai Province (Northwest China) from October 1995 to July 1996. There are no *in situ* measurements of tropospheric ozone profiles in South China. Such measurements are needed to understand the seasonal behavior of tropospheric ozone over this region.

Hong Kong's location (22.2°N, 114.3°E) off the south coast of China puts it under the influence of the Asian monsoon system (Fig. 1). It is also located between the continent of Asia and the ocean (South China Sea and North Pacific Ocean) and is therefore in a zone where different types of air masses meet. It is at the center of the Southeast Asia region where the economy and industry have been developing rapidly in the past 10 years. There are abundant trace gases emitted by anthropogenic and natural sources, both locally and within the subtropical region. The ozone data presented here are expected to help in our understanding of the tropospheric ozone behavior over this tropical–subtropical, monsoonal and coastal region.

2. FIELD STUDY

The first ozone sounding was recorded in Hong Kong on 4 March 1993 on a trial basis (Shun and Leung, 1993). The ECC ozonesondes were launched once a month from October 1993 to August 1995 (with some intensive flights between February and April 1994 as a part of the PEM-WEST B activity), and at least once a week thereafter at the Royal Observatory Hong Kong (ROHK)'s meteorological station at King's Park, an urban site roughly 66 m above mean sea level. Relatively intensive soundings were made during January, March and April 1996, with 9, 9 and 19 soundings, respectively. Less intensive soundings were made through the end of August 1996. The ozone soundings were regularly performed at 06 GMT.

The ozonesonde used consists of the electrochemical concentration cell (the ozone sensor), a nonreactive air pump, an electronic interface board, and

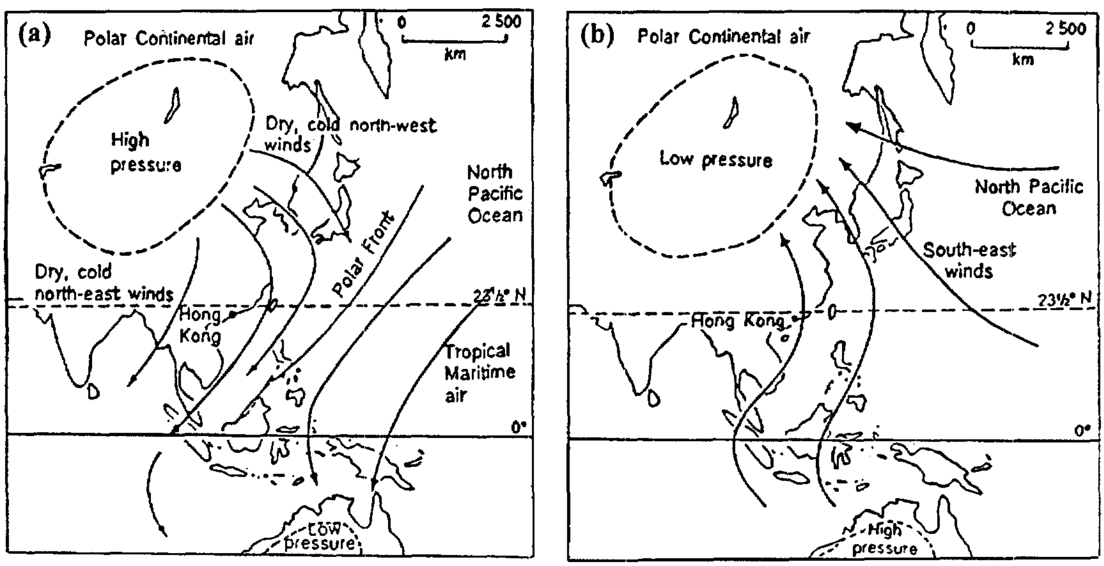

Fig. 1. Pressure and wind systems of winter monsoon (a) and summer monsoon (b) over East Asia (Lau, 1991).

a model RS80 403 MHz Vaisala radiosonde. The ECC sensor (Komhyr, 1969) has been widely used and extensively tested and compared with other techniques (Komhyr *et al.*, 1995). Since ozone is measured quantitatively by the ECC ozonesonde, it is not necessary to normalize the integrated ozone profile amount to an independent measure of column ozone such as the Dobson–Brewer spectrophotometer (Oltmans *et al.*, 1997), although total column ozone measured by the Brewer spectrophotometer is available to us since the beginning of 1995 and provides an important check on the sonde performance. The instrument package is flown with 1500 or 3000 g rubber balloons that normally attain maximum altitudes in excess of 30 km within 1.5–2 h. The attained average balloon ascent rate is about 300 m min^{-1}. Ozone, sonde pump temperature, air pressure, air temperature, humidity and wind data were telemetered back to a ground receiving system (DigiCORA/MARWIN system by Vaisala) and recorded.

3. RESULT AND DISCUSSION

The atmospheric trajectories used in this paper are isentropic, 10 day backtrajectories calculated from the trajectory model of Harris and Kahl (1994) which uses gridded analysis produced by the European Center for Medium Range Weather Forecasts (ECMWF). Any trajectory given by this model is expected to be reasonably representative of the large-scale circulation and may be used to suggest potential source regions, although a specific air parcel's origin cannot be determined exactly.

Results from the measurement of tropospheric ozone profiles over Hong Kong during the PEM-WEST B period (February–March, 1994) have been reported by Yeung *et al.* (1996). This paper looks at the seasonal behavior of tropospheric ozone and some features in tropospheric ozone profiles over Hong Kong. Although ozonesonde data are limited to the past three years, these data are expected to contribute much to the base of our understanding of tropospheric ozone behavior over this region where such data are limited.

3.1. *Seasonal behavior of tropospheric ozone mixing ratio*

The time–height cross section is usually used to show the change of vertical ozone distribution with time. It is constructed here to show the general characteristics of ozone mixing ratio in the whole troposphere. The time–height cross sections of ozone mixing ratio displayed in Fig. 2 summarize the composite profile information during the years of 1993 (October–December), 1994, 1995 and 1996 (January–August), with a total of 4, 19, 22 and 61 soundings, respectively. The vertical profiles of ozone have been averaged every kilometre. The tropopause height over Hong Kong is about 16–18 km. We also plotted the time–height cross sections of tropospheric ozone mixing ratio in individual years of 1994 and 1995 (which are not shown here), and they had a very similar seasonal variation. Although there were limited ozonesonde data in both years, the similarity of these two years of ozonesonde data demonstrates that the overall seasonal variation of tropospheric ozone has been captured.

The most obvious feature is the high ozone mixing ratio throughout the troposphere in springtime, and low ozone mixing ratio below the middle troposphere in summertime. Autumn and winter seem to be the transitional periods. The spring maximum in ozone concentration appears to occur first in the upper troposphere and to propagate down to the lower troposphere. In early summer this ozone maximum begins to decrease from the lower troposphere to the middle troposphere. In the very upper troposphere (from about

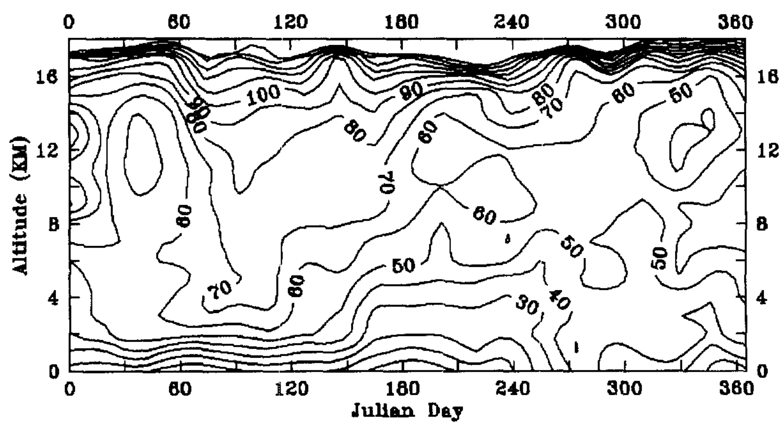

Fig. 2. The composite time-height cross section of tropospheric ozone mixing ratio (ppbv) at Hong Kong, October 1993 to August 1996.

15 km to the tropopause), there seems to be smaller relative seasonal variation in ozone concentration, with a broad maximum of ozone in spring and summer.

The spring maximum in the troposphere is a common feature at many locations in the Northern Hemisphere. Although there has been a long debate over whether the spring maximum is caused by photochemical production from anthropogenic emissions of precursors or intrusion of stratospheric air with a high concentration of ozone (Danielsen, 1968; Fishman and Crutzen, 1978; Liu *et al.*, 1987), our analysis of several high-ozone events in the lower troposphere over Hong Kong suggests to us that those peaks are mostly photochemically produced in the lower troposphere over this region, and that these enhancements make a significant contribution to the ozone budget of the lower troposphere. The initial appearance of the spring maximum at the highest tropospheric altitudes suggests that mixing downward of ozone from the stratosphere also contributes to this maximum. The summer minimum found in this analysis is not particular to Hong Kong and has been observed at many locations, e.g. Cape Kennedy (28.5°N) (Chatfield and Harrison, 1977), lower latitudes over Japan (Logan, 1985; Ogawa and Miyata, 1985). The latter authors attributed the minimum to the increase in photochemical destruction of ozone or to the weak transport of ozone from the stratosphere. In the case of Hong Kong, considering that it is at a lower latitude where the direct influence of stratospheric intrusions is less and it is under the control of southwest summer monsoon most of time, the summer minimum in the lower troposphere over Hong Kong is believed to be due to the increase in photochemical ozone destruction. The southwest summer monsoon brings relatively clean air with high concentrations of water vapor from the South China Sea or the tropical Pacific to the Hong Kong region. High concentrations of water vapor and strong UV radiation in summer may increase the photochemical destruction of ozone. This also implies that photochemical production is not a primary factor in controlling ozone in the lower troposphere over Hong Kong in summer.

The seasonal variation of ozone is strong throughout the troposphere. However, there appears to be a difference in the seasonal variation between the lower and upper troposphere. Although ozone mixing ratio has a minimum in summertime in the lower troposphere, ozone mixing ratio in the upper troposphere tends to have its minimum in late autumn and winter (Fig. 2). This relative minimum in the upper-tropospheric ozone in late autumn and winter can be explained by the East Asia local Hadley circulation, which is discussed in Section 3.4.

3.2. *Seasonal behavior of ozone mixing ratio below 2 km*

The ozone concentration near the ground (0–2 km) can be affected by large local or regional emissions of hydrocarbons and nitric oxides in the presence of sunlight and the resulting photochemical reactions. It may have a more direct effect on the health of human beings. Due to the specific meteorological conditions at these altitudes and the pollutant emission situation in the local area, the ozone-mixing ratio below 2 km may have a very different seasonal behavior from that above.

Ozone profile information below 2 km is summarized in Fig. 3. Individual vertical profiles of ozone have been averaged every 50 m. Very low ozone concentration is found near the surface (below about 500 m) and this may be due to titration by nitric oxide from heavy local traffic as proposed by Yeung *et al.* (1996). Seasonally, there seem to be bimodal ozone peaks in spring and autumn, with even a bit higher ozone in autumn, and a significant minimum in summer. Individual time–height cross-section plots of ozone mixing ratio below 2 km in 1994 and 1995 show

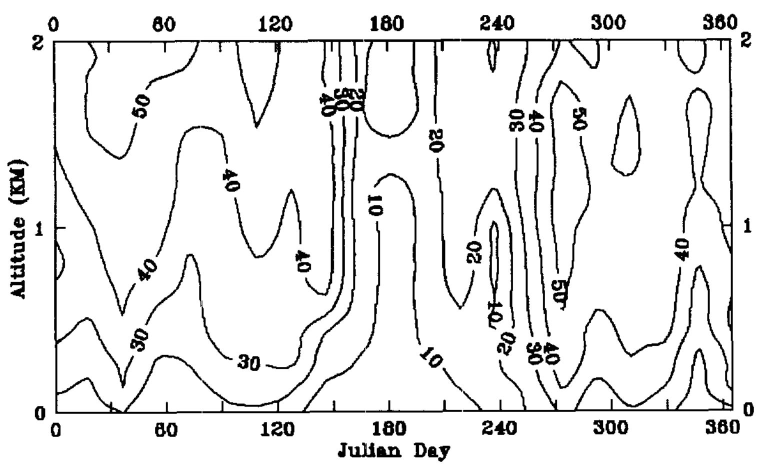

Fig. 3. Same as Fig. 1, except for ozone mixing ratio below 2 km.

that ozone below 2 km in autumn was higher than that of spring in 1994, while they have similar magnitude in 1995. This seasonal pattern agrees with both urban and rural surface ozone observation at Hong Kong. Thus, the behavior of ozone near the surface may not represent that of ozone at higher altitudes. The local meteorological conditions are thought to contribute to this seasonal pattern of ozone-mixing ratio below 2 km.

Chin (1986) presents a summary of climate and weather of Hong Kong. Spring is a transitional season and is marked by very changeable weather. During this period the cool easterlies are displaced by warm southerly winds from time to time. The synoptic pattern undergoes a considerable change from that of the winter season. Cold surges from the north become indistinct and less frequent and the incursions of warm southerly air cause widespread fog or very low-stratus cloud, which usually persists. This cloudy and humid weather in spring may depress the photochemical production of ozone in the lower troposphere near the ground, and also the temperature inversion associated with the persistent low-stratus cloud may keep ozone at higher altitudes from being transported to lower altitudes. Consequently, the ozone near the ground does not show an obvious maximum in spring which appears at higher altitudes.

In summer a large semi-permanent depression develops over the southwestern provinces of China and the prevailing wind is the southerly monsoon (Fig. 1b) which is frequently interrupted by tropical cyclones. Warm, damp air is brought to Hong Kong and the neighboring area from the far south. More than 80% of the annual rainfall occurs during the summer months from May to September inclusive. The summertime minimum (about 10–20 ppbv) of ozone near the ground is ascribed to the air masses of marine origin which are brought by the southwest summer monsoon or tropical cyclones and are relatively clean. As mentioned above, there is also an increase in the photochemical destruction of ozone in the lower troposphere at this time of the year.

During September the prevailing wind shifts rapidly to a mean easterly direction and the temperature begins to fall. Rain also ceases abruptly at the end of the month. The autumn transition period is usually dry and has an abundance of sunshine. The additional sunshine hours may increase the photochemical production of ozone in the lower troposphere near the surface. On the other hand, pressure begins to rise over the continent of China and the northeast monsoon becomes well marked by the end of September, causing cooler air from the northeast or north to sweep southwards. Thus, we suggest that both ample sunshine hours plus air masses carrying precursor pollutants from the northeast or north contribute to the autumn ozone peak in the lower troposphere near the ground.

During winter a cool wind of polar origin, generally known as the northeast monsoon, blows over the coastal regions of China and the China Seas (Fig. 1a). The depth of the monsoon winds is about 2500 m in November but decreases to about 1500 m in February. Above this layer westerly winds prevail. Throughout the season, surface winds are predominantly between north and east. Although many air masses from the north have enhanced precursor pollutants, winter ozone is lower possibly due to weak solar radiation.

3.3. *Seasonal variation of tropospheric ozone column*

In order to integrate the tropospheric ozone profile measured by ozonesonde into the tropospheric ozone column, the tropopause height must first be defined. We use the definition of tropopause defined by WMO (1957). In this paper tropospheric ozone column refers to the ozone column below the first tropopause height.

$$X_t(\mathrm{DU}) = 7.895 \times 10^3 \int_{p2}^{p1} p3(\mathrm{Pa})\,\mathrm{d}(\ln P),$$

where X_t is tropospheric ozone column in Dobson Unit (DU); $p3$ is ozone partial pressure of the layers from the surface to the tropopause in Pascal (Pa); P is air pressure of the corresponding layers; $p1$ is air pressure at ground level and $p2$ is air pressure at tropopause height.

The tropospheric ozone columns determined by integrating all the tropospheric ozone profiles are shown in Fig. 4. Generally, the tropospheric ozone column reaches its maximum (about 40–70 DU) in spring and minimum (about 25 DU) in summer. There is a factor of about 2 difference between minimum and maximum values.

In the Pacific Exploratory Mission over the western Pacific (PEM-West A) (September–October, 1991), Gregory *et al.* (1996) provided insight into the relative importance of lower- and mid-tropospheric ozone (surface-to-9 km altitude) to observed variations in atmospheric total ozone column and pointed out its importance to the interpretation of total column measurements, especially for tropospheric studies. The total ozone column is obtained by integrating the ozonesonde profiles from the surface up to the balloon-burst altitude and determining the amount of residual ozone above the burst altitude using the SBUV ozone climatology data (McPeters *et al.*, 1997). Figure 5 shows the fraction of tropospheric ozone column to the total ozone column over the past three years at Hong Kong. It is seen that the fraction has a maximum (about 15–20%) in springtime and a minimum in summertime (about 9–13%), which closely follows the seasonal variation pattern of tropospheric ozone column. Thus, we conclude that tropospheric ozone behavior contributes much to the seasonal variation of the total ozone column at Hong Kong. The variability of the tropospheric ozone column over the year may not be ignored in the study of total column ozone over this region.

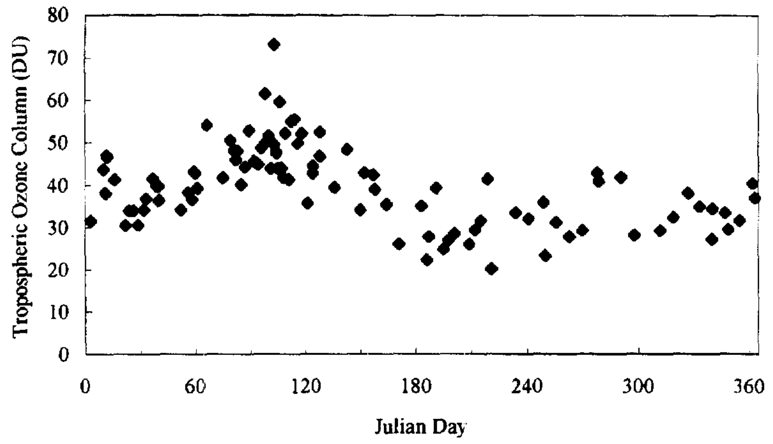

Fig. 4. The composite seasonal variation pattern of tropospheric ozone column, October 1993 to August 1996.

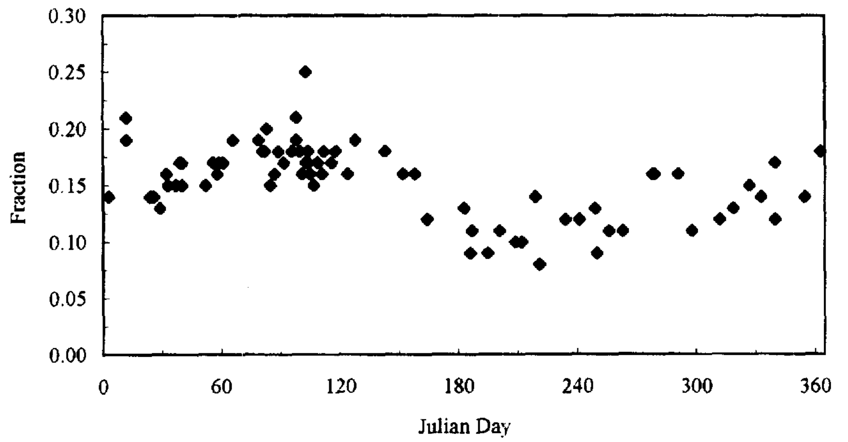

Fig. 5. Same as Fig. 4, except for the fraction of tropospheric ozone column to total ozone column.

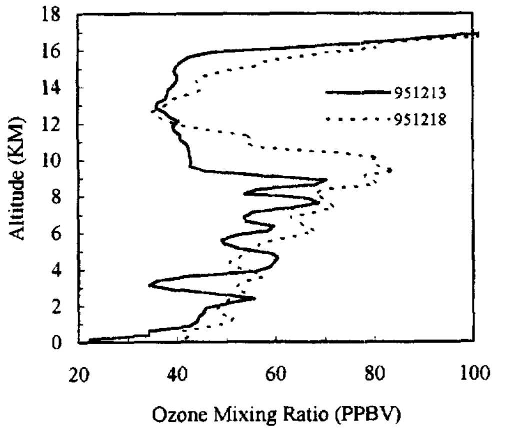

Fig. 6. Representative ozonesonde profiles (951213, 951228) with a relative minimum (< 40 ppbv) of ozone mixing ratio in the upper troposphere during December 1995. 951213 stands for December 13, 1995.

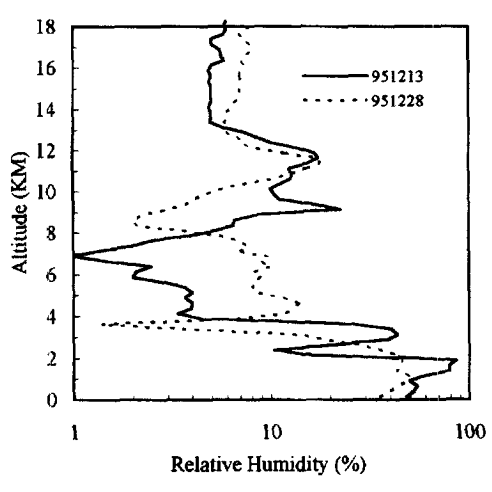

Fig. 7. Relative humidity profiles (951213, 951228) corresponding to ozone profiles of Fig. 6. 951213 stands for 13 December 1995.

3.4. *Ozone in the upper troposphere in late autumn and winter*

The ECC ozonesondes were launched once a week from September to December in 1995 and twice a week from January to early February in 1996. A frequently observed feature in these data is a relative minimum in ozone mixing ratio (as low as 30–40 ppbv) in the upper troposphere. The first profile of this kind was observed early on 25 October 1995. The more usual profiles with increasing ozone mixing ratio with height were obtained before 25 October 1995 and after mid-February 1996. During the period from 10 to 19 January there were also four profiles with higher ozone in the upper troposphere. Figure 6 displays two of the four profiles of tropospheric ozone mixing ratio observed in December 1995, which have been averaged vertically every 250 m. The height range of low ozone mixing ratio in the upper troposphere varies from about 9 to 16 km. Below the relative minimum we observe higher ozone with an increasing trend from the lower to the middle troposphere. The corresponding vertical profiles of relative humidity are shown in Fig. 7. There is enhanced water

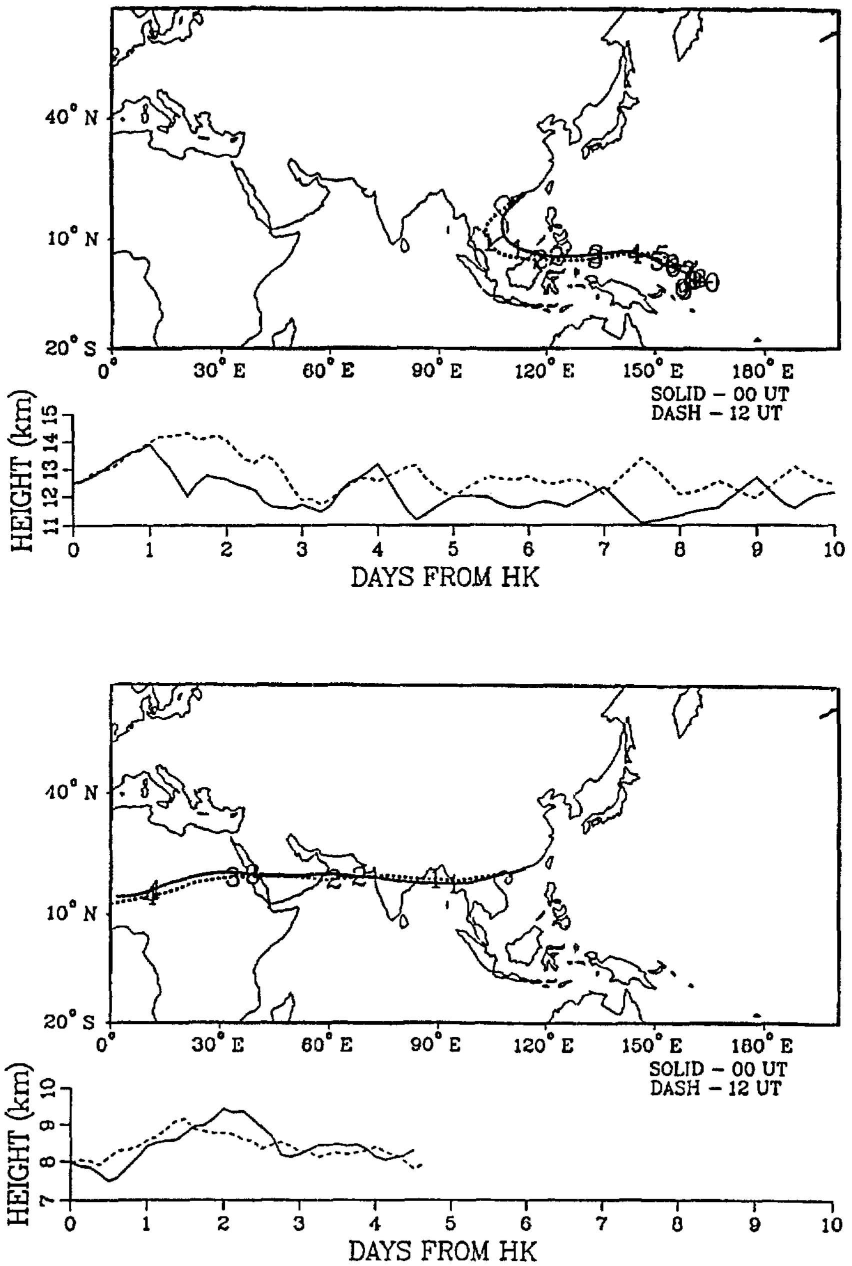

Fig. 8. Isentropic 10 dayback trajectories arriving at 12.5 km (top) and 8 km (bottom) on 28 December 1995.

vapor at the bottom of the low ozone strata. It suggests to us that air masses with low ozone may be of tropical origin where air masses with more water vapor and lower ozone are expected. As mentioned above, this relative minimum of ozone in the upper troposphere is responsible for the seasonal variation of ozone in the upper troposphere, giving the upper-tropospheric ozone the lowest values in late autumn and winter.

Isentropic backtrajectories arriving at 12.5 and 8.0 km for the four ozone profiles during this period are calculated. All the trajectories at the level of 12.5 km show air masses coming from the lower latitude, tropical region. In contrast, air parcels at 8.0 km are from the west, as is illustrated by an example on 28 September 1995 (Fig. 8).

Shown in Fig. 9 are two vertical ozone profiles which are representative of changes in tropospheric ozone during the period from mid-January to early February 1996. There was a dramatic change from higher (960116 profile) to lower (960209 profile) ozone value in the upper troposphere after about 19 January. This relative minimum of ozone remained for more than two weeks. Trajectories arriving at 13.0 km show that the air masses with high ozone are from the west at similar latitude and low ozone is again associated with air masses from the low-latitude tropical region (Fig. 10). It is proposed that the East Asia local Hadley circulation is responsible for this feature in the upper-tropospheric ozone as explained below.

The winter monsoon in East Asia is one of the distinct features of winter circulation over the northern hemisphere (Ding, 1994). During winter, East Asia is dominated by the strong and steady northwesterly and northeasterly monsoon (Fig. 1a). Low-level convergence is concentrated in the vicinity of the cyclonic center located along the northern coast of the island of Borneo, which is also the location of the strongest upper-level divergence in the global wintertime mean flow. Periods of intensification of the winter monsoon, referred to as cold surges, have been shown to be accompanied by an enhancement of the East Asia local Hadley circulation (Chang and Lau, 1980). The upper-level branch of this local Hadley circulation induces a divergent flow in the upper troposphere and may bring convection-processed air masses from the lower latitudes with low ozone northward to the Hong Kong region. The changes between low and high ozone in the upper troposphere are thought to be related to the changes in the East Asia local Hadley circulation, however, this explanation needs further investigation.

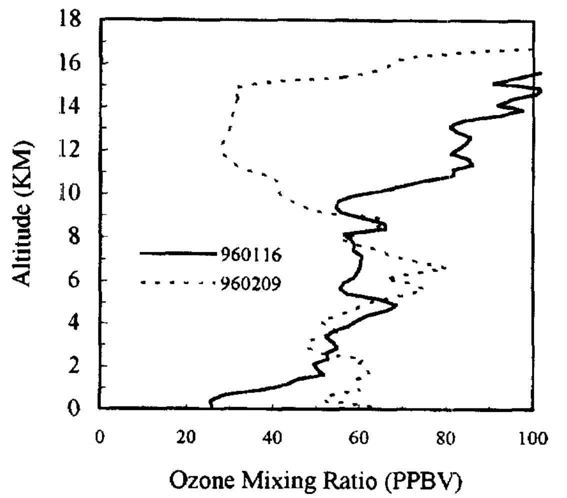

Fig. 9. Representative vertical profiles of ozone mixing ratio between mid-January (960116) and early February (960209) 1996. 960116 stands for 16 January 1996.

4. CONCLUSIONS

Analysis of ozonesonde data from October 1993 to August 1996 has revealed the seasonal cycle in tropospheric ozone distribution over Hong Kong. We conclude that: The tropospheric ozone column has a marked maximum in spring and a minimum in summer. The maximum in spring is a common feature at many locations in the Northern Hemisphere. The minimum in summer is attributed to the onset of the summer monsoon when air flow from the Asian continent is replaced by air from the South China Sea or the tropical Pacific. The tropospheric ozone column has an important influence on the seasonal cycle of the total ozone column at Hong Kong. The variability of tropospheric ozone column over the year may not be ignored in the study of total column ozone. The ozone mixing ratio below 2 km is bimodal with ozone peaks in spring and autumn, thought to be caused by the local meteorological conditions. A frequently observed feature in late autumn and winter is a relative minimum of ozone mixing ratio (as low as 30–40 ppbv) in the upper troposphere (from about 9 to 16 km). Trajectory analysis shows this relative ozone minimum is associated with air masses from the tropical region. This feature is explainable by the East Asia local Hadley circulation.

Acknowledgements—This project is funded by the Research Grants Council of Hong Kong and the Hong Kong Polytechnic University in collaboration with the Royal Observatory Hong Kong (ROHK). The authors greatly appreciate the provision of the monthly ozone data by ROHK and the help given by ROHK's personnel at King's Park meteorological station in launching of ozonesondes. Special thanks are due to Dr. W. L. Chang of ROHK for his coordination of the project and his many helpful comments and Prof. Xiaoyan Tang of Peking University for her support of this research project. Ozonesondes launched during the PEM-WEST B experiment period were provided by NASA.

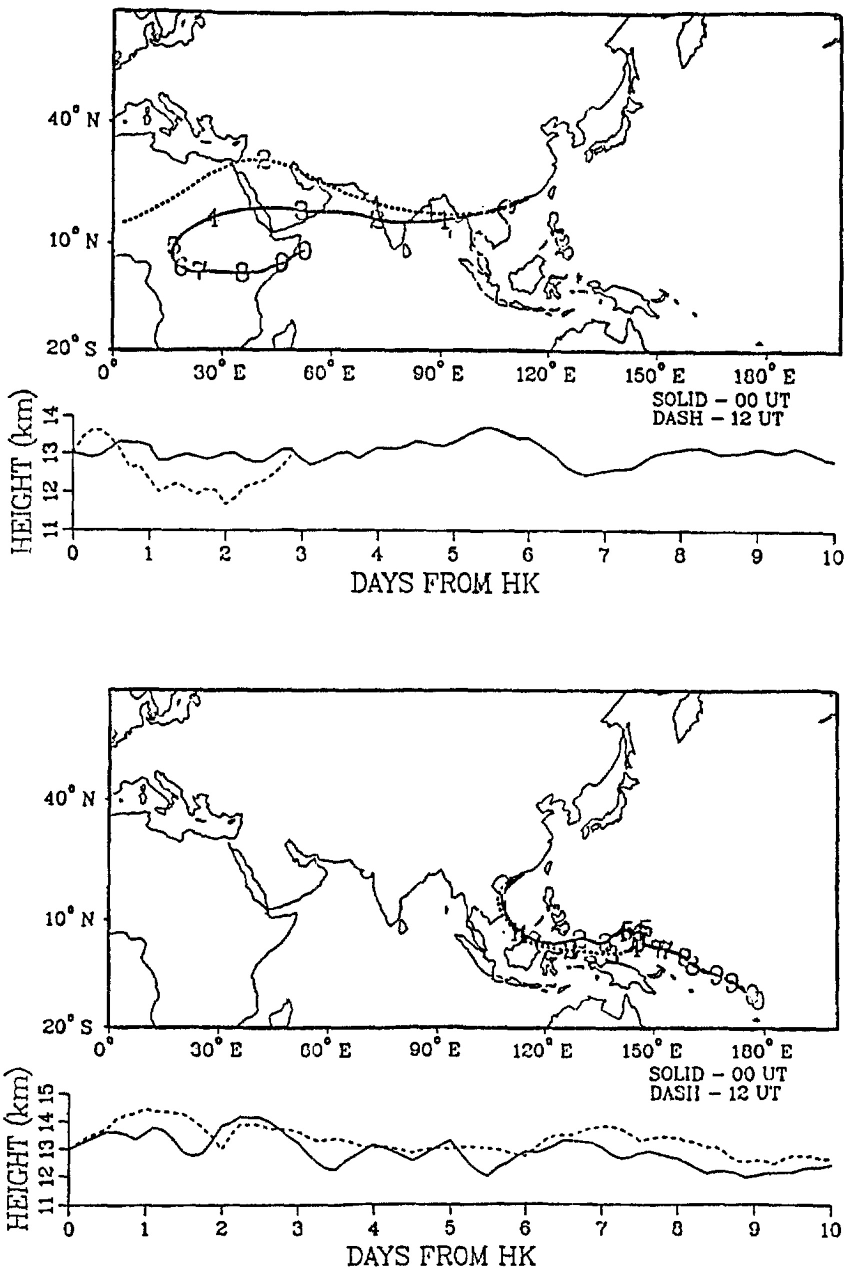

Fig. 10. Isentropic 10 day backtrajectories arriving at 13 km on 16 January (top) and 9 February (bottom) 1996.

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