



## On springtime high ozone events in the lower troposphere from Southeast Asian biomass burning

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### Abstract

Ozone peaks with mixing ratios as high as 138 ppbv were observed in the lower troposphere (2.5–4.5 km) over Hong Kong in spring. Simultaneously observed high humidity suggests that this enhanced ozone was not the result of transport from the upper troposphere. Back trajectory analysis suggests that these enhancements resulted from lateral transport. Air masses arriving at the altitude of the ozone peaks appear to have passed over continental Southeast Asia where the bulk of biomass burning occurs at this time of the year (February–April). We hypothesize that biomass burning in this region provided the necessary precursors for the observed ozone enhancement. As far as we know this is the first observation of highly enhanced ozone layers associated with biomass burning in continental Southeast Asia. © 1999 Elsevier Science Ltd. All rights reserved.

*Keywords:* Tropospheric ozone; Vertical profiles; Hong Kong; Continental Southeast Asia; Back trajectory analysis

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### 1. Introduction

Tropospheric ozone plays a significant role in controlling the chemical composition, air quality, and climate evolution of the troposphere (Fishman et al., 1979; WMO, 1994; Mohnen et al., 1995; IPCC, 1995). Balloon sounding has been widely used to measure ozone in the free troposphere and the stratosphere, however, most ozonesonde stations are located in the middle or high latitudes. The World Meteorological Organization (WMO) has recommended that balloon soundings of the vertical distribution of ozone be made especially in the tropics

where such data are scarce (WMO, 1993). In response to this recommendation, the Hong Kong Observatory (HKO) began making routine ozone profile soundings in October 1993, both to gather such data from the South China Sea area and as part of its contribution to WMO's Global Atmosphere Watch (GAW) program. Normally these soundings are made once a month due to cost considerations and the personnel resources that are required. During NASA's Pacific Exploratory Mission West-B (PEM-West B) mission to Hong Kong in February and March 1994 and between September 1995 and April 1997 in collaboration with Hong Kong Polytechnic University, weekly (and occasionally more frequent) soundings were carried out. A description of the PEM-West B mission can be found in Hoell et al. (1997).

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It was found that in spring profiles sometimes displayed layers of high concentration in the lower troposphere at elevations from 2.5 to 4.5 km. The highest mixing ratio observed was 138 ppbv. This exceeded the 60–80 ppbv reported for the lower troposphere over Japan (Tsutsumi and Makino, 1995) and the 112 ppbv observed during the biomass burning season in Brazil (Kirchhoff and Marinho, 1994). The highest amount observed over Hong Kong was comparable to the 140 ppbv reported for a pollution event over the eastern US (Johnson and Viezee, 1981). In these studies, the enhancements in ozone were attributed to either downward transport from the stratosphere or photochemical production in the troposphere.

The paper documents several high ozone events observed in the lower troposphere over Hong Kong. Back trajectory analysis was used to help identify the source of these enhanced concentrations. A brief introduction of the field study is given in Section 2. Section 3 describes the observed springtime high ozone events and the associated transport features revealed by the trajectory analysis. The origin of ozone enhancement is discussed in Section 4. Finally, conclusions are given in Section 5.

## 2. Field study

Ozone profiles over Hong Kong (22.2°N, 114.3°E) were obtained using balloon borne ozonesondes and the DigiCORA system manufactured by Vaisala. The ozone sensor in the sonde is of the electrochemical concentration cell (ECC) type, which has been widely used and extensively tested and compared with other techniques (Komhyr et al., 1995). In addition to ozone readings, the sonde also gives simultaneous measurements of pressure, temperature, dew point, and wind. Soundings were normally made at 1400 local time (060UTC) at HKO King's Park Meteorological Station, an urban site roughly 66 m above mean sea level. Stringent calibration procedures were followed before each launch. One measure of the reliability of the results is demonstrated by the agreement between the sounding and those measurements obtained by a DC-8 aircraft deployed to Hong Kong during PEM-West B (Yeung et al., 1996). Ozonesonde data between 1995–1997 were validated by comparison with total ozone measured simultaneously with a Brewer ozone spectrophotometer. Details of the HKO ozonesonde sounding operation can be found in Shun and Leung (1993). An analysis of the seasonal behavior of tropospheric ozone at Hong Kong using parts of this data set appears in Chan et al. (1998).

## 3. Observed ozone peaks in the lower troposphere and trajectory analysis

We first describe the observed enhanced ozone events in the lower troposphere and then use trajectory analysis

to reveal transport features associated with air masses at the ozone peaks. The trajectories used here are isentropic 10-day back trajectories and the trajectory model (Harris and Kahl, 1994) used gridded analyses produced by the European Centre for Medium Range Weather Forecasts (ECMWF). Such trajectories represent the large-scale circulation and may be used to suggest potential source regions, though the origin of a specific air parcel cannot be determined exactly.

### 3.1. February 1994

This first instance of enhanced ozone concentrations in the lower troposphere over Hong Kong was observed during PEM-West B's expedition to Hong Kong from 21–28 February 1994 (Yeung et al., 1996). Two representative profiles are adapted here in Fig. 1 (see their Fig. 1 for details). On February 21 the ozone profile showed a minor peak at altitude 2.7 km with mixing ratio of about 65 ppbv. Two days later this peak became very sharp and had a mixing ratio of 95 ppbv at 2.6 km. On the 25th the peak appeared at a somewhat higher altitude of 3.2 km with a mixing ratio of 89 ppbv. The peak cannot be distinguished in the profile on the 27th. Above these peaks the ozone mixing ratio gradually decreased to about 50 ppbv and remained constant up to 6–8 km. Simultaneous measurements made by DC-8 showed high carbon monoxide concentrations and suggest that the air masses associated with enhanced ozone concentrations on this occasion were not of stratospheric origin (Yeung et al., 1996).

We used trajectory analysis to investigate the suggestion of Yeung et al. (1996) that the ozone-rich air was due to lateral transport. Back trajectories were calculated at the altitudes of 1, 3, and 5 km. At 1 km wind speeds were

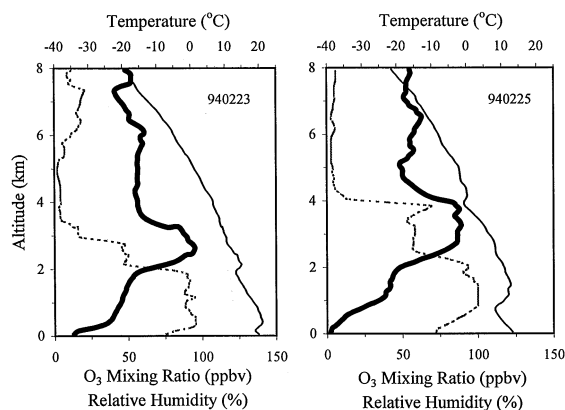


Fig. 1. Representative vertical profiles of ozone mixing ratio (ppbv, thick solid line), temperature (°C, thin solid line) and relative humidity (% , dashed line) during 21–28 February 1994, in the lower troposphere over Hong Kong (Yeung et al., 1996). 940223 stands for 23 February 1994, and so on.

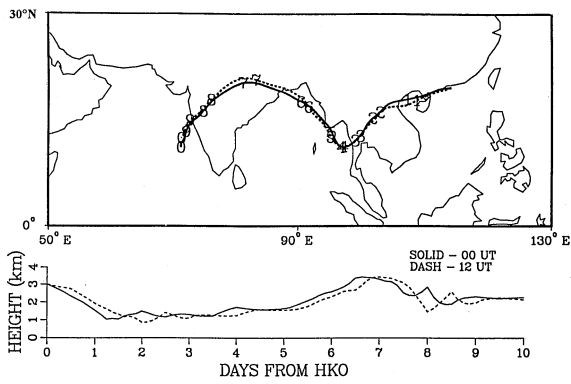


Fig. 2. Isentropic 10-day back trajectory arriving at 3.0 km on 25 February 1994.

light and air parcel origins were confined within a couple of hundred kilometers of Hong Kong. At 5 km flow was strong and mostly from the west. Trajectories for 23 February show air parcels originated at lower levels of northern continental Southeast Asia and rose to 3 km

over Hong Kong. At this time, an ozone concentration of about 95 ppbv was observed. From 24–26 February the air flow was traced back to low altitudes over middle continental Southeast Asia. On 25 February, the peak concentration was 89 ppbv. As an example, the trajectory arriving at 3 km on 25 February is given in Fig. 2. On 27 February the ozone peak disappeared when the air flow was much weaker and trajectories show origins near the southeast tip of continental Southeast Asia.

### 3.2. March 1996

A similar event with even higher ozone in the lower troposphere occurred during 15–25 March 1996 (Fig. 3). On 15 March an ozone peak at 3.3 km had a mixing ratio of about 81 ppbv. Four days later on 19 March, the ozone mixing ratio at 3.0 km increased to 110 ppbv. Thereafter, ozonesondes were launched every other day in order to see how this high ozone event evolved. The ozone profile on 21 March still showed a very sharp peak at 3.8 km with an even higher value of 118 ppbv. Laminar structures appeared near the ozone peak (about

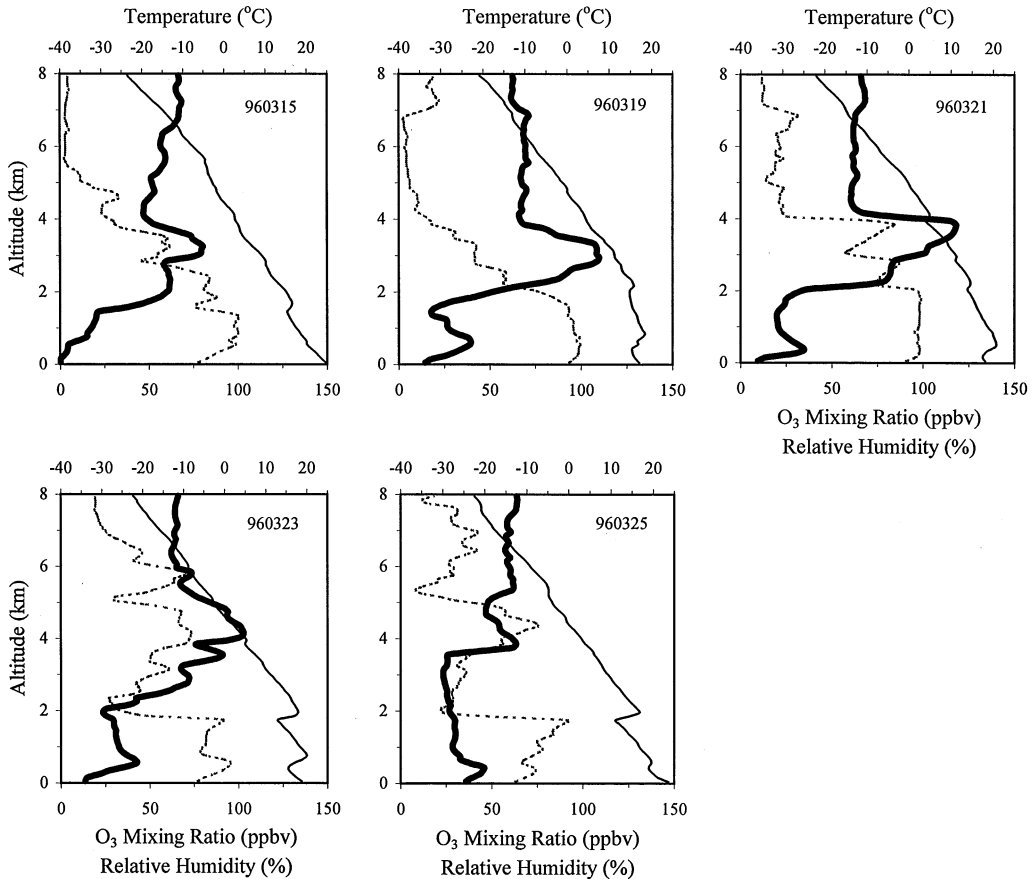


Fig. 3. Same as Fig. 1, except for the period of 15–25 March 1996.

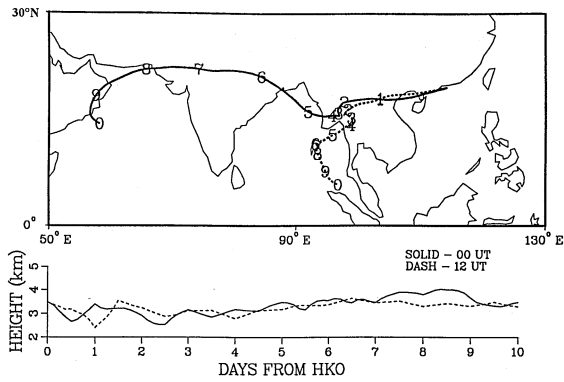


Fig. 4. Isentropic 10-day back trajectory arriving at 3.5 km on 21 March 1996.

4.0 km) in the ozone profile on 23 March, but the peak concentration was still as high as 103 ppbv. By 25 March the large peak had disappeared.

Trajectories arriving at the altitude of the ozone peak during this event showed most of the air masses passed over continental Southeast Asia at lower levels 1–3 days

before ascending to 3.5 km over Hong Kong. As an example, the trajectory arriving at 3.5 km on 21 March is presented in Fig. 4. On 23 March, possibly due to the shift in air flow from westerly to southeasterly, the ozone peaks broadened but persisted around 4.0 km. Trajectories were traced back to the southeast marine area, passing over the Philippines. By 25 March, the peaks had disappeared with transport from the southeast.

3.3. April 1996

The third highest ozone event in the lower troposphere appeared during 7–16 April 1996 (Fig. 5). Although no obvious peak near 2–4 km can be seen in the ozone profile measured on 5 April, on 7 April a very large peak appeared at 2.5–3.5 km with a concentration of 128 ppbv. From 9 April on, ozonesondes were launched daily until 18 April. During the period from 9–12 April, the peak decreased somewhat and remained at the altitude of 3.0–3.5 km with concentration about 90–100 ppbv. On 13 and 14 April, the ozone profiles showed less pronounced ozone peaks of 70–80 ppbv. However, the strongest ozone event was observed on 15 April. This

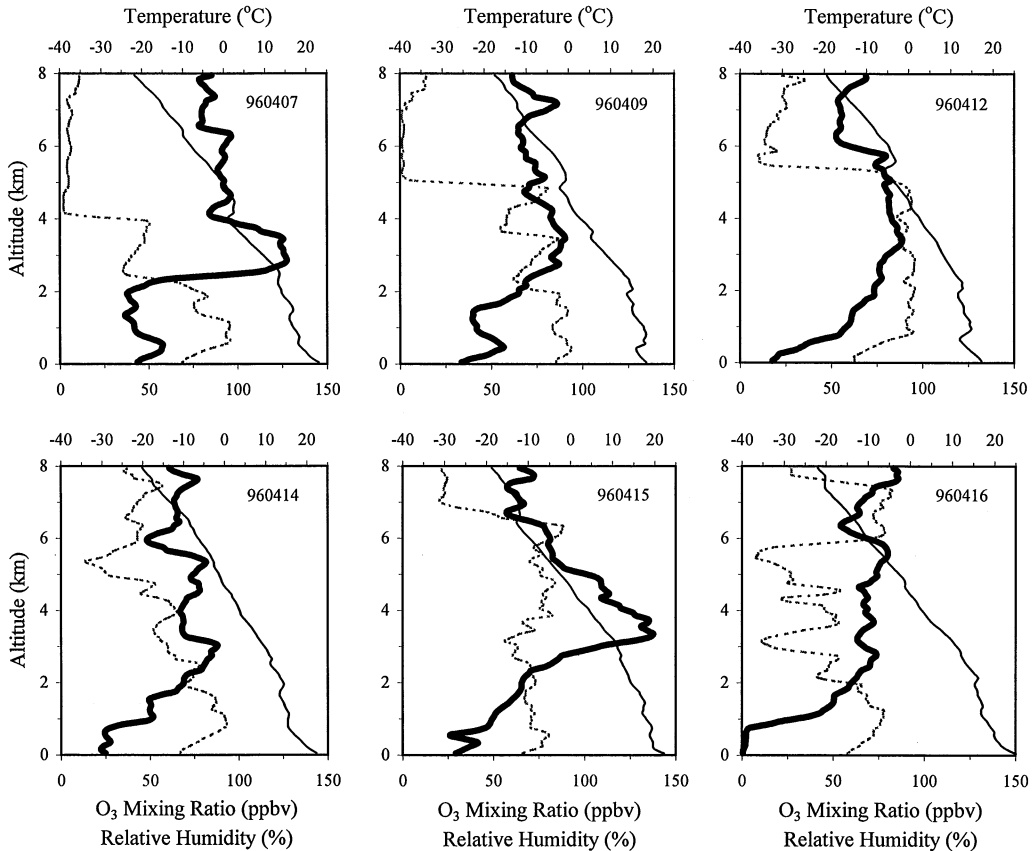


Fig. 5. Same as Fig. 1, except for the period of 7–16 April 1996.

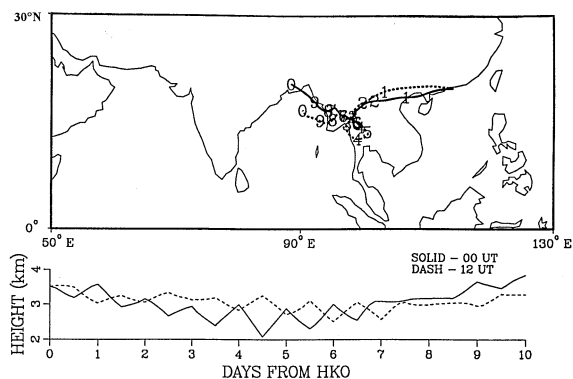


Fig. 6. Isentropic 10-day back trajectory arriving at 3.5 km on 15 April 1996.

event had a concentration of 138 ppbv at 3.3 km. The concentration at the peak decreased to about 70 ppbv and the peak was not well defined on the next day, 16 April.

Most trajectories at the altitude of 3.5 km in this event are similar to the preceding events, with air masses passing over continental Southeast Asia. The trajectory for the day with the largest peak value, 15 April, is shown in Fig. 6.

#### 4. Possible origin of ozone enhancement

The magnitude, frequency of occurrence, and the persistence of the layers of high ozone concentration over Hong Kong as well as the potential impact of such events on the regional ozone distribution warrant further investigation into the source of these layers. Several candidates suggest themselves as potential sources of the enhanced ozone. Among these are local photochemical production, mixing downward and lateral transport from a source in the upper troposphere or perhaps the stratosphere, or transport of ozone and its precursors from a very photochemically active source region. Both the meteorological information obtained with the ozone profile and the air parcel trajectory information are key in distinguishing among these possibilities.

The ozone profiles were obtained near the center of a heavily urbanized area and in a region of southeastern China which is experiencing explosive industrial growth and is likely a significant source of ozone precursor emissions. The proximity of the observatory to central Hong Kong actually produces depressed ozone values in the boundary layer on many occasions because of ozone titration by recently emitted nitric oxide (NO). This can be seen in several of the profiles (Figs. 1, 3, and 5). The trajectories are also very consistent in showing that air parcel origins are not southeastern China, but rather they skirt the southern coastal area.

Both the meteorological information obtained with the ozone profiles and the trajectories are consistent in showing that the layers of high ozone mixing ratio do not have their origin in the upper troposphere or stratosphere. The humidity in the enhanced ozone layers remains quite high within the layer, generally exceeding 50%. There is a dramatic decrease in humidity above the top of these layers. This is in contrast to the humidity profiles seen during transport from the upper troposphere or stratosphere at other subtropical sites (Oltmans et al., 1996). The trajectories also show little evidence of strong descent of air parcels found in the enhanced ozone layers, which is also in contrast to what was observed in our previous studies (Oltmans et al., 1996).

This leaves the case for a strong photochemical source not associated with the rapidly industrializing, urban centers of China. Spring in Hong Kong is a transitional season when the synoptic pattern undergoes a considerable change away 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 and is accompanied by a temperature inversion (Chin, 1986). Occasionally there exist multiple temperature inversions in the lower troposphere, as seen from temperature profiles (Figs. 1, 3, and 5). Smoke plumes with high ozone and its precursors can be trapped by such temperature inversions, where they are inhibited from being transported and mixed vertically, thus producing well-defined layers of enhanced ozone. The inversion at the bottom of the layer also suggests that the high concentrations were not due to mixing upwards from the local boundary layer. Air parcel trajectories are extremely consistent in showing arrivals from continental Southeast Asia within the previous 2–3 days (Figs. 2, 4, and 6). All of the trajectories also indicate some rising motion during the last two days of air parcel travel time to Hong Kong. Because air parcel transport from northern Southeast Asia is quite rapid, photochemical production time is limited. This suggests that ozone is primarily generated in the source region prior to transport over Hong Kong. Alternately, the photochemical production en route may be unusually large. In many of the cases, as mentioned above, it appears that there may be an underlying cloud layer (Chin, 1986) as the plume passes over Hong Kong based on the near 100% humidity found in the lowest 2 km in almost all of the profiles. Such a cloud layer provides a very reflective surface with greater ultraviolet irradiance and thus more ozone generation from precursor emissions.

NO<sub>x</sub> emissions from fossil fuel combustion and industrial activities in continental Southeast Asia are much lower than in many of the Pacific Rim countries (Akimoto and Narita, 1994). Therefore, it does not seem likely that industrial emissions are the primary source of ozone

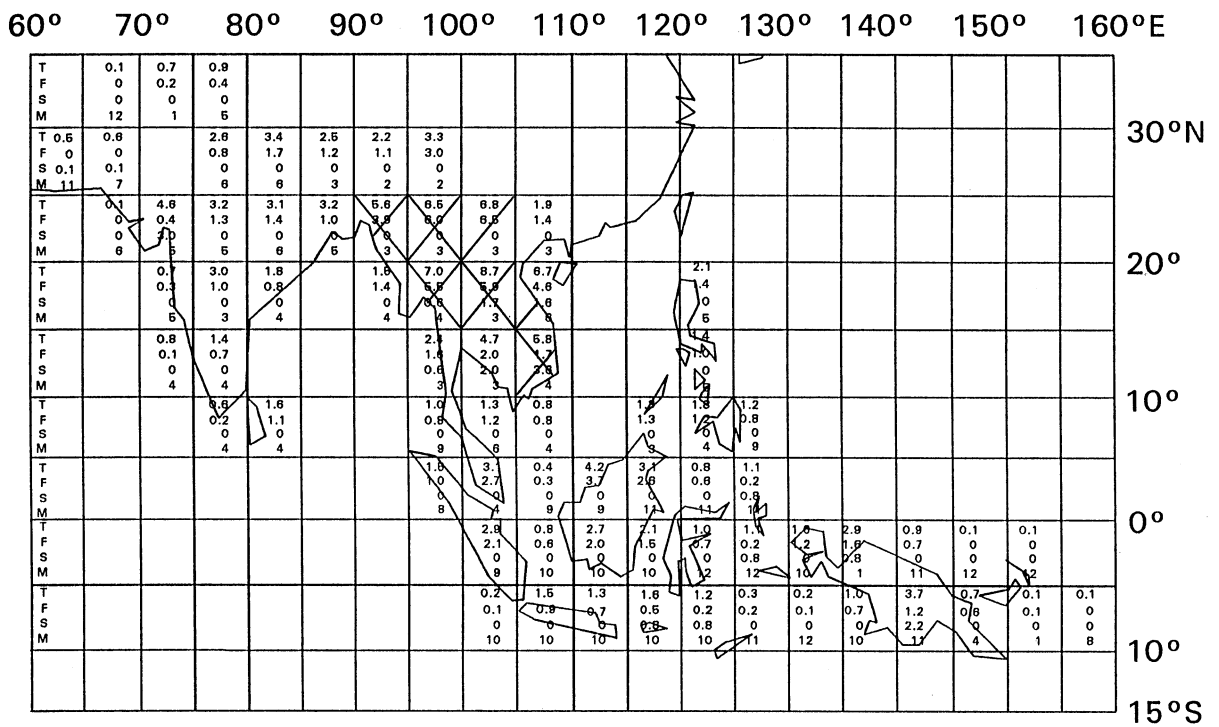


Fig. 7. Distribution of total amount of biomass burned  $T$ , amount burned in forest fires  $F$ , amount burned in savanna fires  $S$ , and amount of fuel wood and agricultural residuals burned ( $T - (F + S)$ ) in tropical Asia in the fourth month  $M$  (usually the maximum month) of the burning season during the late 1970s (in teragrams of dry weight per month; adapted from Hao and Liu, 1994). (Cross-hatching indicates areas where total burned biomass exceeds 5 Tg of dry weight per month.)

precursors responsible for the observed peaks. A more likely candidate for ozone precursors and the subsequent ozone production is the considerable biomass burning that occurs over continental Southeast Asia at this time of year. Biomass burning, which has been investigated primarily in the Amazon region of South America and over southern Africa, is a significant source of  $\text{NO}_x$ ,  $\text{CO}$ , and hydrocarbons necessary for ozone production (Crutzen et al., 1979; Delany et al., 1985; Crutzen and Andreae, 1990).

Although tropical Asia is a region of extensive biomass burning (Hao and Liu, 1994; Molingreu et al., 1996; Jones, 1997), there are only very limited reports of ozone enhancements associated with this burning. What reports there are have been for Indonesia (Komala et al., 1996; Folkens et al., 1997) and New Guinea (Kim and Newchurch, 1998). As far as we know this is the first attempt to link observations of enhanced ozone amounts with biomass burning over continental Southeast Asia (Burma, Thailand, Laos, Cambodia, and Vietnam). At present the identification of fire data from satellite images for individual days or even composites for individual months is a very time consuming process. Such information is not available except for limited time periods and for particular geographic regions. Although we cannot

make a direct link to particular fires burning during the 2 years for which we have ozone profiles, we can show that there is regular and extensive burning in the postulated source region at the time of year when the enhanced ozone layers were observed.

Hao and Liu (1994) have produced a climatology of tropical biomass burning of various types and the time of the maximum burning. While burning in Indonesia and New Guinea peaks late in the year, the burning in continental Southeast Asia is most intense in March and April (Fig. 7, adapted from Hao and Liu, 1994). The amount of biomass burned in all tropical Asia is quite large. It is about one-half of the amount burned in tropical America, and about one-third of that in tropical Africa. Elvidge and Baugh (1996) conducted a survey of biomass burning in India and Southeast Asia during 1987, using nighttime images from the Defense Meteorological Satellite Program operational system, and found the fires were primarily in Thailand, Burma, Laos, Cambodia, and Vietnam during February and March. Recently, Jones (1997) studied the distribution of vegetation fire during the dry season of 1992/93 in continental Southeast Asia using AVHRR satellite data. He found that 70% of fires occur within forest ecosystems, which account for 55% of the land surface of the region. Most fires were found to

occur in the last 2 months (March, April) of the season (Stott, 1988). Non-forest fires, which may be associated with crop residue burning, grassland fires, and savanna woodland fires, occur during the early and middle parts of the dry season (November–February).

Because the layers of enhanced ozone generally have their bases at about 2 km and are about 2–3 km thick, only weak vertical motion is required to mix ozone and/or its precursors from the boundary layer into the layer that is transported over Hong Kong. As noted earlier the back trajectory calculations show air parcel ascent in the final 2 days of travel from the northern section of continental Southeast Asia. This is consistent with a low altitude source for these layers.

## 5. Conclusions

We report observations of greatly enhanced ozone concentrations at elevations of 2–5 km over Hong Kong. These layers had mixing ratios often exceeding 100 ppbv and occurred during February through April of 1994–1996. During 1997 the observed peak value only reached about 80 ppbv. We also show that over continental Southeast Asia at this time of year there is extensive biomass burning that is a likely source of abundant ozone precursors and subsequent photochemical ozone production. The back trajectories clearly indicate that the air parcels in the enhanced ozone layers over Hong Kong passed over the region of intensive seasonal burning. The trajectory analyses also show that air parcels rise from lower levels in the final two days of travel from the postulated source region. This is consistent with the high humidity found in the layers, which indicates that the layers have their origin in the lower troposphere rather than higher altitudes. Because data on fires during individual months is not available for Southeast Asia during the time of the ozone profiles, we were not able to connect the enhanced layers with regions of burning on a case-by-case basis. In the future, if such information does become available, we plan to expand the study to try to make a more direct link to biomass burning in this region. To our knowledge burning in continental Southeast Asia has not previously been studied as far as its impact on the ozone distribution though the amount of material that is burned each year appears to be quite significant. From the results presented here it appears likely that ozone produced in connection with burning in this region affects the ozone distribution in the lower troposphere downwind of the burning in a substantial way.

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