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Increasing surface ozone concentrations in the background atmosphere of southern China, 1994–2007

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Abstract

Tropospheric ozone is of great importance with regard to air quality, atmospheric chemistry, and climate change. In this paper we report the first continuous record of surface ozone in the background atmosphere of South China. The data were obtained from 1994 to 2007 at a coastal site in Hong Kong, which is strongly influenced by the outflow of Asian continental air during the winter and the inflow of maritime air from the subtropics in the summer. Overall, the ozone concentration increased by an averaged rate of 0.55 ppbv/yr, with a larger increase in autumn (0.68 ppbv/yr). We also examine the trend in air masses from various source regions in Asia. Using local wind and concurrently measured carbon monoxide (CO) data to filter out local emissions, the mean ozone in air masses from eastern China, using the pooled averaging method, increased by 0.64 ppbv/yr, while ozone levels in other air-mass groups showed a positive trend (0.29–0.67 ppbv/yr) but with lower levels of statistical significance. An examination of the nitrogen dioxide (NO_2) column concentration data obtained from GOME and SCIAMACHY reveals an increase in atmospheric NO_2 in the three fastest developing coastal regions of China, whereas NO_2 in other parts of Asia decreased during the same period. It is believed that the observed increase in background ozone in Hong Kong is primarily due to the increased emissions of NO_2 (and possibly volatile organic compounds (VOCs) as well) in the upwind coastal regions of mainland China, which is supported by the observed positive CO trend (5.23 ppbv/yr) at the site. The increase in background ozone contributed two thirds of the annual increase in "total ozone" in the downwind urban areas of Hong Kong, suggesting the need to consider distant sources when developing long-term strategies to mitigate local ozone pollution, although short-term strategies should be aimed at sources in Hong Kong and the adjacent Pearl River Delta.

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1 Introduction

Ozone is a key constituent in the troposphere and plays a role in air quality, atmospheric oxidizing capacity, and climate change. At high concentrations, ozone has detrimental effects on human health, crops, and vegetation (NRC, 1991; Chameides et al., 1999). Ozone regulates the oxidizing capacity of the atmosphere via the production of hydroxyl radicals, which are the principal cleansing reagents in the atmosphere. Ozone is also a greenhouse gas that directly contributes to global warming (IPCC, 2007). Therefore, long-term ozone trends and the underlying cause(s) comprise an area of extensive research worldwide (Vingarzan, 2004; Oltmans et al., 2006; IPCC, 2007, and references cited therein).

Tropospheric ozone is determined by downward transport from the stratosphere (Danielsen, 1968), deposition loss to the Earth's surface, and photochemistry in the troposphere involving hydrocarbons, carbon monoxide (CO), and nitrogen oxides (NO_x) (Crutzen, 1973). Because of the complex interactions of chemical and meteorological factors, the trends in tropospheric ozone vary both in terms of sign and magnitude and possible causes in different regions of the world (Naja and Akimoto, 2004; Oltmans et al., 2006; IPCC, 2007). In the mid-latitudes of Europe and North America and in Japan, tropospheric ozone increased significantly during the 1970s and 1980s, but more recently it has leveled off and in some places declined. The trends in ground-level ozone in the urban and rural areas of industrialized regions are strongly linked to the changes in anthropogenic emissions of ozone precursors (Fiore et al., 1998; USEPA, 2008). In remote areas, changes in emissions in distant source regions, wild biomass burning, and atmospheric circulation all play an important role in ozone trends. For example, several studies have attributed the increase in tropospheric ozone in the remote areas of the western United States to rising emissions in Asia via trans-Pacific transport (Jacob et al., 1999; Parrish et al., 2004; Jaffe and Ray, 2007). The enhanced transport of ozone from the stratosphere due to climate change is thought to have increased the ozone levels in the upper troposphere in mid- to high-latitude regions (Collins et al.,

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2003).

China has undergone rapid urbanization and industrial development in the past three decades. A key question is how the anticipated sharp rise in pollutant emissions affects the atmospheric composition of chemically and radiatively important trace gases and aerosols. Although intensive field studies reveal the frequent occurrence of ground-level ozone pollution in photochemically active seasons in suburban and rural areas of East China (e.g., Cheung and Wang, 2001; Gao et al., 2005; Shao et al., 2006; Wang et al., 2006a,b), knowledge of the long-term ozone trends in China is very limited because of the lack of long-term continuous data from regionally representative sites. Ding et al. (2008) analyzed aircraft ozone data obtained from the MOZAIC (Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft) program for the 1995–2005 period (with data concentrating in 1997 to 1998 and 2005) and found that summertime ozone in the boundary layer near Beijing had increased by about 2% per year. Xu et al. (2008) found a positive trend in daytime ozone at a polluted rural site in the Yangtze River Delta, based on data collected during seven measurement campaigns lasting 3–15 months in the 1991–2006 period. In the maritime regions impacted by Asian continental outflow, Chou et al. (2006) reported a positive trend in surface ozone in northern Taiwan for the 1994–2003 period, whereas the ozone data obtained at several Japanese sites indicated no obvious trends during the 1990s in regionally polluted air masses from China (Naja and Akimoto, 2004).

In this report, we present the first long-term and continuous record of surface ozone obtained at a non-urban site from 1994 to 2007 in southern China. The ozone data were collected at a regional background air monitoring station of The Hong Kong Polytechnic University at Hok Tsui, Hong Kong. Influenced by Asian monsoons, the site receives pollutant-laden continental flow in the winter and clean maritime air from the South China Sea in the summer, making it a suitable location to monitor the background ozone of subtropical eastern Asia. First shown are the overall trend, seasonal cycles, and trends in four seasons. Then a cluster analysis of back trajectories over the 14 years is conducted to identify the major types of air-mass groups, and the ozone

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trend in each group is examined. Also analyzed are the trends in the satellite-derived nitrogen dioxide (NO_2) columns in different source regions in Asia and the trends in surface CO observed at Hok Tsui. Finally, the impact of the background air on the local ozone trends in urban Hong Kong is assessed.

2 Data and methodologies

2.1 Study site and instrumentation

Hong Kong Polytechnic University's background air monitoring station was established at Hok Tsui to monitor the changes in the composition of background air and to investigate the chemistry and transport of air pollution in subtropical Asia (http://www.cse.polyu.edu.hk/~cetwang/_Researches/Guangdong/HokTsui/index.htm). The station is an integral part of the PEM-West, TRACE-P, and ACE-Asia campaigns (Wang et al., 1997; Lam et al., 2001; Wang et al., 2003; Cohen et al., 2004), and has been involved in other intensive studies (see the website for a full list of publications). The reader is referred to these publications for a more detailed description of the site. Briefly, the site is located at the southeastern tip of Hong Kong Island (22.217° N, 114.25° E, 60 m above sea level), on a cliff with a 270° view of the South China Sea. Most of the time, the station is upwind of the urban areas of Hong Kong and the adjacent Pearl River Delta (PRD) in Guangdong province.

Measurement instruments are housed in a temperature-controlled container. Ambient air samples are drawn through a perfluoroalkoxy (PFA) Teflon tube (outside diameter, 12.7 mm; inside diameter, 9.6 mm), which is connected to a PFA manifold. The length of the sampling tube was 17 m (14 m after July 2007). The inlet of the sample line was located 12 m above the rooftop of the laboratory (9 m after July 2007). A bypass pump draws air at a rate of 15 L/min. The residence time of air in the sample line and manifold was 4.2 s (3.4 s after July 2007). An inline Teflon filter (Fluoroware Inc., Chaska, Minnesota) is placed upstream of each analyzer to prevent particles from

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entering the analyzer.

Ozone is measured with a commercial UV photometric analyzer (Thermo Environment Instrument Inc. (TEI), Models 49, 49C, and 49i). Zero checks are performed every day by automatically injecting charcoal-scrubbed air (Wang et al., 1997; Lam et al., 2001). The ozone analyzer is calibrated by a UV photometric calibrator (TEI Model 49PS) approximately every six months. The ozone standard has been cross-checked against two National Institute of Standards and Technology (NIST) primary ozone standards maintained by the Hong Kong Environmental Protection Department and National Institute for Environmental Studies of Japan (Tanimoto et al., 2007). Excellent agreement (difference <1%) was obtained.

To facilitate the analysis of ozone trends, the CO data concurrently measured at the site is also utilized. CO is measured with a gas filter correlation, non-dispersive infrared absorption instrument (TEI Model 48 before March 2006 and Teledyne API Model 300 thereafter). The reader is referred to Wang et al. (1997), Lam et al. (2001), and Wang et al. (2001) for detailed descriptions of the two instruments. Both analyzers have an internal heated catalyst to convert CO to CO_2 while allowing other gases and water vapor to pass through. Zero checks are conducted every two hours for a period of 15 min. The analyzer is calibrated against an NIST-traceable standard. Both analyzers have been compared with the canister-based sampling method and gas chromatographic analysis technique, with good agreement (within 10%) achieved.

2.2 Satellite data

Satellite data was used for an examination of the spatial distribution of and trends in NO_2, a precursor of photochemically produced ozone. The monthly mean global tropospheric NO_2 columns were used, with 0.5 degree resolution, obtained from GOME (Global Ozone Monitoring Experiment) for the 1995–2002 period and SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) from 2003 onwards. The data were obtained via TEMIS (Tropospheric Emission Monitoring Internet Service; http://www.temis.nl/index.html).

2.3 Back trajectories and cluster analysis

To examine the origin and transport pathways of the large-scale air masses arriving at the study site, Ten-day back trajectories were calculated for each hour using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model Version 4.8 (Draxler and Hess, 1998). The meteorological data used to drive the model were the 6-hourly Air Resources Laboratory (ARL) FNL archive data with a resolution of 190 km for the 1997–2006 period (ftp://www.ready.noaa.gov/pub/archives/fnl), and the 2.5°×2.5° NCEP/NCAR Reanalysis data (ftp://www.ready.noaa.gov/pub/archives/reanalysis) were used in lieu of the missing data in the former source in 1994–1997 and in 2007. The endpoint of the trajectories was 300 m above ground level at the Hok Tsui site, which is in the middle of the marine boundary layer. Cluster analysis (Dorling et al., 1992) was then used to group trajectories into clusters and determine the optimum number of clusters. The results are shown in Sect. 3.2.1.

3 Results and discussion

3.1 Overall trend and seasonal pattern of ozone

Figure 1 shows the monthly averaged ozone over the 1994–2007 period and the regression line of the monthly data. Large seasonal and inter-annual variation is clearly seen. Overall, there is a statistically significant ($p<0.01$) upward trend in ozone during the 14-year period, with a mean rate of 0.55 ppbv/yr. Figure 2 shows the composite seasonal pattern of ozone. A similar seasonal cycle has previously been reported based on the data from shorter periods (1–3 years) (Lam et al., 2001; Wang et al., 2005), and can generally be explained by Asian monsoon circulation. The dominance of marine air masses, coupled with rainy and unstable weather, leads to low levels of ozone (and other trace gases) in summer. Pollution-laden continental flow from the north, in conjunction with the stable and warm weather in autumn, contributes to the

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ozone maximum in autumn (winter for primary pollutants such as CO). These 14-year data give a climatologically average maximum of 47 ppbv in October and minimum of 18 ppbv in July.

Figure 3 shows the seasonal ozone averages and the regression results for the 14-year period. Winter includes December, January, and February; spring, March, April, and May; summer, June, July and August; and autumn, September, October, and November. The mean ozone increased by 0.68 ppbv/yr in autumn, with lower rates (0.50, 0.41 and 0.52) in winter, spring and summer, respectively. The autumn data dominate the overall trend result.

The ozone trends in the four seasons at Hok Tsui are somewhat different from those at another coastal site, WanLi, in northern Taiwan during 1994–2003 (Chou et al., 2006). At the latter site, which is about 800 km northeast of Hong Kong, the largest rate of increase of ozone occurred in spring (0.71 ppbv/yr, p=0.0044), followed by summer (0.55 ppbv/yr, p=0.1467), autumn (0.15 ppbv/yr, p=0.7073), and winter (0.10 ppbv/yr, p=0.7738) (see Table 1 of the reference). In contrast, coastal Hong Kong saw the largest rate of increase in autumn, and the autumn trend had the highest level of statistical significance. In comparison with the ozone trends derived from the limited data in source regions in eastern China, the overall trend in Hong Kong was similar to that in the planetary boundary layer over Beijing (Ding et al., 2008). At a polluted rural site, Lin'an, in the Yangtze River Delta, surface ozone showed a larger rate of increase in daytime and a decrease at night, with no obvious overall trend (Xu et al., 2008). The opposite trend in the daytime and nighttime data at Lin'an is attributed to the increase in NO_x emissions in the region leading to enhanced photochemical production of ozone in the daytime and the titration of ozone at night (Xu et al., 2008). At Hok Tsui, ozone increased both in the daytime and the nighttime (figure not shown), which indicates that the site is not significantly impacted by local pollution sources.

3.2 Climatology of large-scale transport

3.2.1 Transport pattern using cluster analysis

To discover the origins and transport pathways of air masses in the background atmosphere of southern China, the results of cluster analysis of the hourly trajectories for the 1994–2007 period were examined. Cluster analysis yielded a total of seven air-mass groups. The two marine sub-groups arriving from the southwest and southeast were grouped into one "Marine" group, and three continental air-mass groups into one "East China" group, in part because the concentrations of ozone and CO were similar in the subgroups. Figure 4 shows the four main groups and the percentage occurrence of each, with shaded areas as the mean NO_2 column concentration obtained from GOME and SCIAMACHY for the 1996–2007 period. The four air-mass groups are: "Marine", "Central China+PRD", "East China", and "Aged continental".

The Marine group originated from southeastern Asia and passed over the South China Sea before arriving at the site. This group, which can be considered background air in the South China Sea, mainly (68%) occurred in summer (June, July, and August) and accounted for 30% of all trajectories. The Aged continental air mass originated in the Asian continent, spent a long time over the East China Sea, and approached Hong Kong from the east. This air mass accounted for 13% of total trajectories. The East China group originated in central Asia, and passed over the highly industrialized coastal regions of eastern China. This group accounted for 45% of total trajectories, making it the predominant air-mass group. The Central China+PRD air mass passed over central China, the PRD, and Hong Kong. The two latter air-mass groups mainly occurred in autumn and winter (79% and 68% of the time, respectively). The altitudes of these groups are also shown. The three continental air groups originated at altitudes higher than those of the marine group. The 14-year trajectory results confirm that large-scale air masses arriving at Hok Tsui are mainly from the northeast and south, and infrequently come from urban Hong Kong itself or the adjacent PRD.

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3.2.2 The ozone trends in different air-mass groups

Next examined were the ozone trends in the four air-mass groups. As the study site is close to the urban center of Hong Kong and trajectories based on coarse meteorological data may not capture the local to meso-scale dynamic processes that could transport Hong Kong's emissions to the site, we imposed additional constraints when examining the chemical characteristics of air masses in the Marine, East China, and Aged continental groups. The surface winds observed at Waglan Island (WGL) by the Hong Kong Observatory were used. WGL is situated 8 km southeast of Hok Tsui. The following criteria were adopted: the wind speed at WGL must be greater than 2 m/s, and the wind direction in the preceding six hours at WGL must be within 20–250° (i.e., not from urban Hong Kong or the PRD). In addition, for the Marine and Aged continental groups, the concurrently measured CO must be lower than 400 and 500 ppbv, respectively.

Segregating/filtering the ozone data using cluster analysis and the abovementioned criteria led to limited data points distributed unevenly in different seasons and times of the day. As ozone in the lower troposphere has strong seasonal and diurnal variation, a pooled statistical method (Taylor and Cihon, 2004) was adopted to calculate the mean ozone level in each group. The data in air-mass group were further divided into 16 sub-groups (four seasons and four time-periods of a day), and the means and standard deviations in the 16 sub-groups were pooled to obtain the overall mean and standard deviations for that air-mass cluster. It has been shown that the pooled method gives results with higher levels of statistical significance (Ding et al., 2008).

The means and standard deviations obtained using the pooled method for the ozone and CO data are shown in Table 1 for the above air-mass groups and with the wind/CO constraints. As expected, the mean CO concentration was the lowest in the Marine group, with a mean value of 170 ppbv (±64 ppbv). The Central China+PRD air mass had the highest CO concentration (473 ppbv±204 ppbv), followed by the East China (327 ppbv±143 ppbv) and Aged Continental (240 ppbv±93 ppbv) groups. The average

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ozone was the lowest in the Marine group (21 ppbv±10 ppbv), also as expected. The highest ozone concentration was found in the East China group (50 ppbv±14 ppbv), with lower concentrations in the Central China+PRD (41±20 ppbv) and Aged continental groups (38±16 ppbv). The chemical data are consistent with the results of air-mass classification using the back trajectories, in terms of the expected concentrations of the two gases.

Table 2 shows the ozone 14 years trend in each air-mass group. There was a positive (0.64 ppbv/yr) trend in ozone in the East China group, with a much higher level of statistical significance (p=0.08) compared with that of the other air-mass groups (p=0.21–0.27). The average growth rate was 0.29, 0.45, and 0.67 ppbv/yr in the Marine, Aged continental, and Central China+PRD groups, respectively. The application of the wind and CO filter had little effect on the trend results (the slope and p value) for the East China group, but made a large difference for the Aged continental and Marine groups.

3.3 Relationship between the ozone trend and anthropogenic emissions in source regions

An important issue is to determine the main cause of the increasing background ozone in coastal south China. Figure 5 shows the trend in tropospheric NO_2 columns derived from satellite data for various parts of Asia which may have affected the ozone concentrations at Hok Tsui. These regions (see also Fig. 4) include: the North China Plain (NCP, 114° E–120° E, 34° N–41° N), the Yangtze River Delta (YRD, 118° E–123° E, 28° N–33° N), the Pearl River Delta (PRD, 112° E–115° E, 21° N–24° N), Korea and Japan (Kr–Jp, 126° E–140° E, 32° N–39° N), Taiwan (TW, 119° E–122° E, 22° N–26° N), and Southeast Asia (SA, 98° E – 108° E, 10° N–22° N).

Similar to the finding of Richter et al. (2005), the satellite data indicate significant NO_2 column increases in the three most economically developed regions of China. The North China Plain (which is home to two megacities, Beijing and Tianjin) had the highest mean NO_2 column concentration and the largest rate of increase, followed by

the Yangtze River Delta (which is home to China's largest city, Shanghai), and the Pearl River Delta (which is the world's major consumer goods manufacturing base). A pronounced seasonal variation in NO_2 columns can be seen, with higher NO_2 in winter due to the longer lifetime of NO_2 and greater NO_x emissions in northern regions (Richter et al., 2005). It is interesting to note that the abnormally high ozone concentrations in 2004 coincided with the highest tropospheric NO_2 columns in the PRD and the NCP for that year. In contrast to the increasing trend in NO_2 in the three regions of China, an overall decreasing trend in NO_2 was observed in Taiwan, Korea and Japan, and Southeast Asia.

These results show that the increasing background ozone in the south China coastal region is associated with the increasing NO_2 column concentration in upwind eastern China. Although other factors such as climate change may be playing a role in the ozone trend, the pronounced increase in NO_x (and possibly, volatile organic compound (VOC)) emissions in eastern China is believed to be the main cause of the increasing ozone levels at Hok Tsui. This notion is supported by the increasing concentration in CO – which is produced mainly from the burning of fossil fuels and biomass and is a precursor of ozone. Figure 6 shows that the mean CO concentration at Hok Tsui increased by 5.23 ppbv/yr during 1994–2007. During the same period, CO emissions from Hong Kong decreased by 55% (http://www.epd.gov.hk/epd/english/environmentinhk/air/data/emission_inve.html). The increase in the CO concentration at Hok Tsui contrasts with the decreasing tropospheric CO observed in the past two decades in many other places worldwide (Novelli et al., 2003, 2007), and provides strong evidence of increased CO emissions (and likely other pollutants) in mainland China.

3.4 The impact of background air on ozone in urban Hong Kong

As for other urban areas, Hong Kong and the adjacent PRD experience serious photochemical ozone pollution. Hourly ozone concentrations of 150–200 ppbv have been observed in photochemical episodes (e.g., Wang et al., 1998; Zhang et al., 2007, 2008).

It is of great interest to see to what extent the change in background ozone has affected the ozone trend in urban Hong Kong. Figure 7 compares the trend of ozone at Hok Tsui, which, as discussed previously, is upwind of the Hong Kong's urban areas most of the time, with that observed at an urban monitoring station (Central/Western (C/W)), which is 14.5 km west of Hok Tsui and operated by the Hong Kong Environmental Protection Department. The ozone concentrations at this site have increased since the early 1980s (Chan et al., 2004; http://www.epd-asg.gov.hk/english/report/files/aqr07e.pdf). The site is in an urban/residential area on the densely populated northern side of Hong Kong Island and is strongly influenced by traffic sources. As ozone at this site is titrated by freshly emitted NO via $O_3+NO \rightarrow NO_2+O_2$, we also show O_3+NO_2 to present the overall oxidant abundance in the urban area. This quantity includes the amount of O_3 that is temporally lost due to the NO titration and also the NO_2 that is directly emitted by vehicles and other local sources. Thus the atmospheric concentration of "total ozone", (Total O_3), which is defined as the observed O_3 in the urban areas plus the O_3 that is titrated by NO, can be expressed as:

$$[\text{Total } O_3]=[O_3 + NO_2]_{\text{observed}}-[NO_2]_{\text{directly emitted}}$$

Figure 7 shows that the ozone level at CW increased at an average rate of 0.65 ppbv/yr during the 1994–2007 period, with O_3+NO_2 increasing at a slightly higher rate of 0.69 ppbv/yr. Ambient concentrations of NO_x at CW decreased during the same period at a rate of −0.68 ppbv/yr (p=0.02) (see the lower panel in Fig. 7) because of the implementation of several control measures during the period. As freshly emitted NO_x contains 10–30% NO_2, we estimate the rate of change of NO_2 due to direct emissions as −0.14 ppbv/yr, assuming that 20% of fresh NO_x is in the form of NO_2. Thus, the trend in "total ozone" in the urban area was 0.69−(−0.14)=0.83 ppbv/yr. This implies that the increase in background ozone accounted for 66% of the increase in the total ozone in the urban area of Hong Kong. This finding suggests the importance of taking long-range (super-regional) transport into account in the management strategy of ground-level ozone pollution in the urban areas of Hong Kong and the PRD. However,

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short-term control strategies should aim to reduce sources within Hong Kong and the PRD, which have led to peak concentrations of 150–200 ppbv during episode days.

4 Conclusions

Motivated by the need to understand the atmospheric impact of East Asia's rapid industrialization, a regional air monitoring station was established in the early 1990s in a coastal area of Hong Kong. This paper presents the ozone and CO results obtained from 1994 to 2007. The 14-year data reveal increasing ozone levels at an average rate of 0.55 ppbv/yr. This increase is associated with the increase in tropospheric NO_2 in the fast-developing coastal regions of China and the increase in the concurrently measured CO. These findings, combined with the back trajectory results, suggest that the observed increase in ozone levels is very likely the result of the increase in the emission of ozone precursors in upwind source regions in the eastern coastal regions of China. Long-range transport makes a significant contribution to the increase in "total ozone" in the urban area of Hong Kong, thus suggesting the importance of considering super-regional transport in future ozone control. Although this study reveals an association between the increase in ozone level and the increase in emissions in upwind source regions, modeling studies are needed to confirm this association and to quantify the relative contribution from each source region to the ozone trend.

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Table 1. Pooled means and standard deviations of ozone and CO in the four air-mass groups.

Air mass type	O_3 (ppbv)	CO (ppbv)
East China	50 (14)	327 (143)
Central China+PRD	41 (20)	473 (204)
Aged Continental	38 (16)	240 (93)
Marine	21 (10)	170 (64)

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Table 2. Trend of yearly pooled average of ozone mixing ratios in the four air-mass groups.

Air mass type	With wind/CO filter		Without wind/CO filter	
	O_3 trend (ppbv/yr)	p value	O_3 trend (ppbv/yr)	p value
East China	0.64	0.08	0.61	0.06
Central China+PRD	–	–	0.67	0.21
Aged Continental	0.45	0.21	0.75	0.07
Marine	0.29	0.27	0.45	0.11

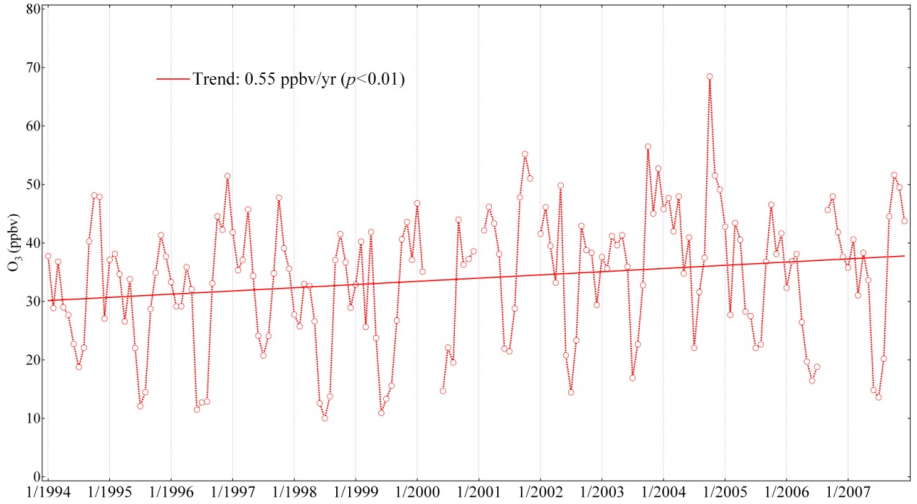

Fig. 1. Monthly mean ozone mixing ratios and the regression line at Hok Tsui, Hong Kong, during 1994–2007.

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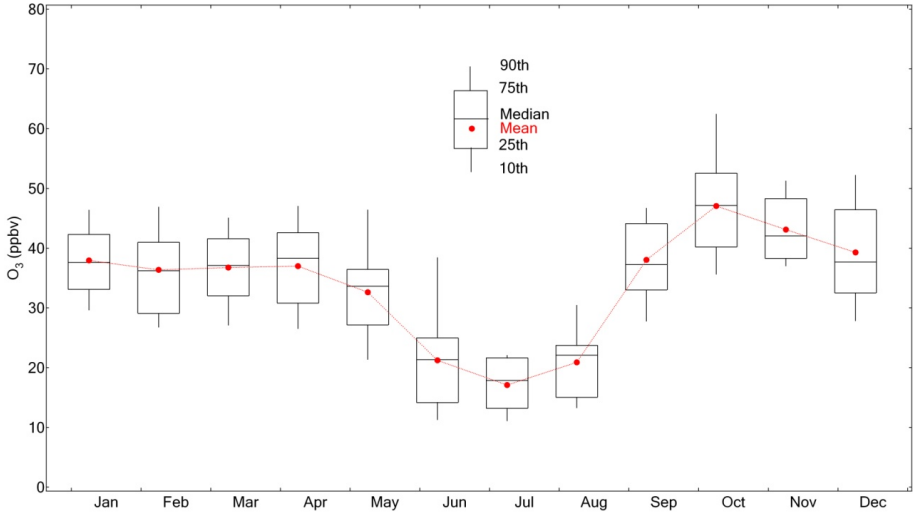

Fig. 2. Whisker plot of the seasonal variation of ozone at Hok Tsui during the 1994–2007 period.

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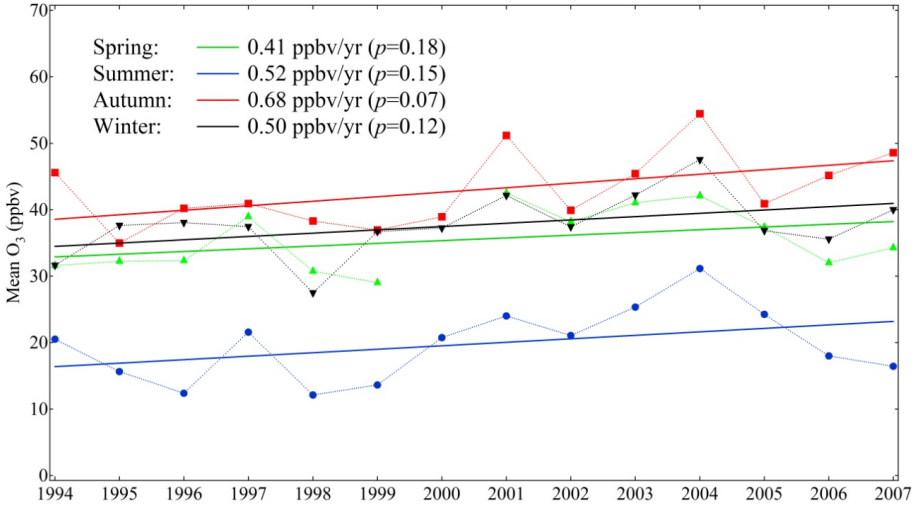

Fig. 3. Seasonal averaged ozone mixing ratios at Hok Tsui and the trend results (spring: March, April, and May; summer: June, July, and August; autumn: September, October, and November; winter: December, January, February).

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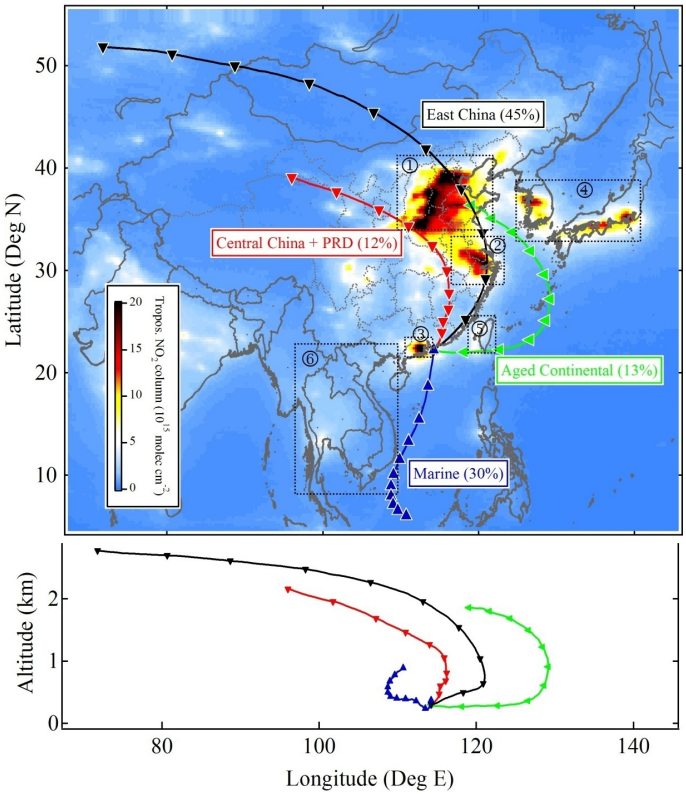

Fig. 4. Four major types of 10-day backward trajectories at Hok Tsui. The percentage of each type during the 1994–2007 period is shown in parentheses. The NO_2 column abundance data from GOME and SCIAMACHY (averaged over 1996–2007) are shown in the shaded areas. The boxes in dashed lines are potential sources for ozone at Hok Tsui: (1) NCP: North China Plain, (2) YRD: Yangtze River Delta, (3) PRD: Pearl River Delta, (4) Kr-Jp: Korea and Japan, (5) TW: Taiwan, (6) SA: Southeast Asia.

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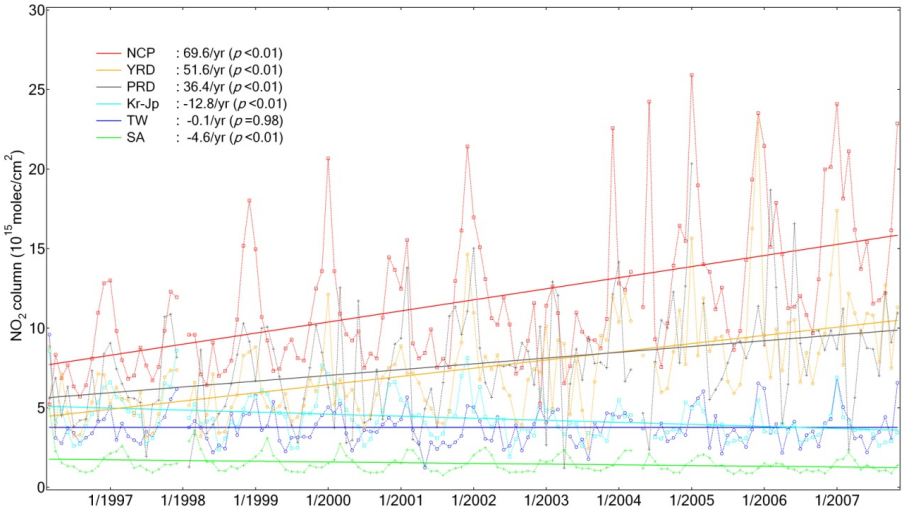

Fig. 5. Trends in the monthly mean tropospheric NO_2 column concentration retrieved from GOME (March 1996–June 2002) and SCIAMACHY (July 2002–November 2007) for NCP, YRD, PRD, Kr-Jp, TW, and SA.

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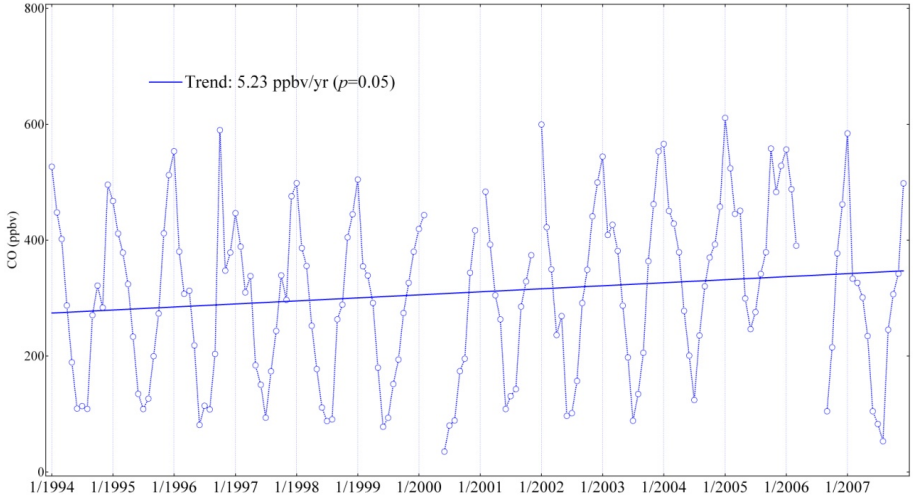

Fig. 6. Monthly mean CO mixing ratios and the regression line at Hok Tsui, Hong Kong, during 1994–2007.

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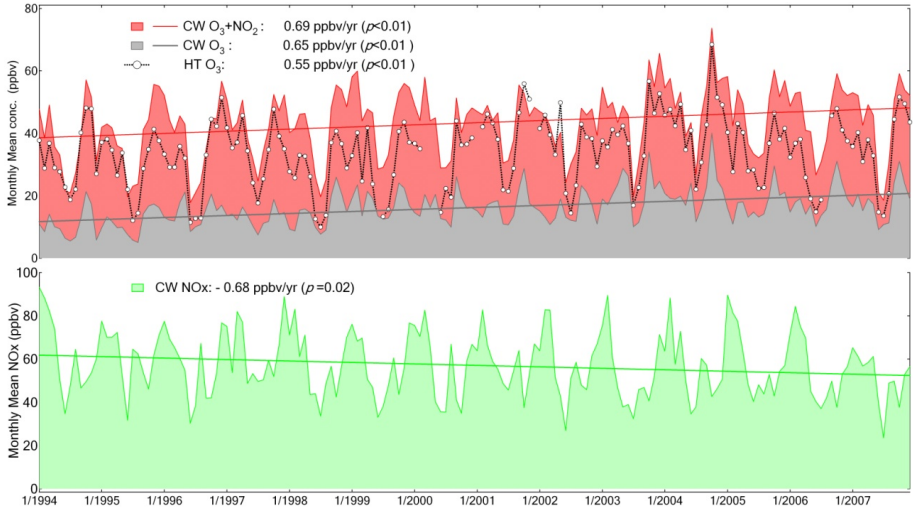

Fig. 7. Monthly mean O_3 mixing ratios at Hok Tsui, O_3 and O_3+NO_2 at an urban site (Central/Western) in Hong Kong, and their regression lines (upper panel); NO_x at Central/Western in Hong Kong and the regression line (lower panel).

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