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Temporal variation of elemental carbon in Guangzhou, China, in summer 2006

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Abstract

In situ measurements of the mass concentration of elemental carbon (EC) and mixing ratios of carbon monoxide (CO) and carbon dioxide (CO₂) were made at Guangzhou, an urban measurement site in the Pearl River Delta (PRD), China, in July 2006. The average±standard deviation (SD) concentrations of EC, CO, and CO₂ were 5 $4.7\pm2.3\mu$ gCm⁻³, 798±459ppbv and 400±13ppmv, respectively. The trends of these species were mainly controlled by synoptic-scale changes in meteorology during the campaign. Based on back trajectories, data are analyzed separately for two different air mass types representing northerly and southerly flows. Northerly air masses, constituting about 25% of the campaign, were mainly impacted by stagnant conditions, 10 resulting in elevated levels of pollutants. On the other hand, southerly air masses measured during most of the campaign were mostly influenced by clean marine air. The diurnal patterns of EC, CO, and CO₂ exhibited peak concentrations during the morning and evening hours coinciding with rush-hour traffic. The diurnal variations of EC and Δ EC/ Δ CO closely followed the traffic pattern of heavy-duty vehicles (HDV) 15 in Guangzhou, similar to that observed in Beijing. The level of EC in this cam-

- paign was similar to values reported during previous studies at other sites surrounding Guangzhou. The average slopes of $\Delta EC/\Delta CO$, $\Delta EC/\Delta CO_2$, and $\Delta CO/\Delta CO_2$ were 0.0054 μ gCm⁻³/ppbv, 0.15 μ gCm⁻³/ppmv, and 46.4 ppbv/ppmv, respectively, agreeing reasonably well with their respective emission ratios derived from regional emission
- ²⁰ ing reasonably well with their respective emission ratios derived from regional emission inventories.

1 Introduction

25

A major portion of the fine-mode aerosols in the urban atmosphere are carbonaceous aerosols. Carbonaceous aerosols are generally classified into organic carbon (OC) and elemental carbon (EC). EC is also referred to as black carbon (BC) and can ab-

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sorb light, therefore acting as a positive radiative forcing agent in the troposphere and causing a negative radiative forcing at the earth surface (Ramanathan et al., 2008; Jacobson et al., 2000; Chuang et al., 2003; Kim et al., 2008). It has been estimated that the global mean clear-sky radiative forcing of BC is about +0.4–0.8 W/m² (IPCC,

5 2001). BC also affects cloud albedo and cloud formation (Conant et al., 2002; Nenes et al., 2002). Incomplete combustion of fossils fuels and biomass burning are the main sources of EC and CO. Quantitative information of their relative emissions can be used to characterize various emission sources (Hansen et al., 1989).

Emissions of various gaseous and particulate pollutants in Asia are increasing due
to rapid industrialization and urban development (Streets et al., 2003). Elevated levels of the various aerosols and gaseous species in East Asian countries are of great concern because of their impacts on the atmospheric environment on regional and continental scales, for instance a reduction in precipitation of 10–20% and increasing tendency of floods and drought (Huang et al., 2007; Menon et al., 2002). In China,
¹⁵ major economic expansion and industrialization are occurring in mega-city clusters like Beijing, Tianjin, Bohai, and the Pearl River Delta (PRD) (Shao et al., 2006). The PRD is a densely industrialized region where emissions from a large number of petrochemical, automobile, and electronics manufacturing units are significant. Guangzhou is the main commercial and industrial city in the PRD region, with an area of 7434.4 km² and

²⁰ population of over 10 million in the year 2006.

Previous studies of anthropogenic aerosols and gaseous pollutants at Guangzhou and the surrounding urban areas mostly focused on the relationships of OC and EC and their seasonal variations (Cao et al., 2003, 2004, 2007; Duan et al., 2007; Ho et al., 2006; Cheung et al., 2005). Some studies have also characterized aerosol chemical composition and optical properties (Andreae et al., 2008; Liu et al., 2008). However, none of the previous studies have reported the relationship of EC to other combustion tracers (e.g., CO, CO₂), particularly in light of their emission inventories from different sources. These relationships can be used to characterize sources and also to validate the existing emission inventories of these species. There is a need to study the diur-

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nal variations of aerosols and gaseous species for their source attributions by making continuous time-resolved measurements.

For the first time, we report the temporal variations of hourly averaged EC, CO, and CO₂ in the PRD region in summer in light of transient meteorological conditions ⁵ and diverse emission sources. We discuss the diurnal patterns of these species and slopes of $\Delta EC/\Delta CO$ in relation to traffic patterns. The estimated slopes of $\Delta EC/\Delta CO$, $\Delta EC/\Delta CO_2$, and $\Delta CO/\Delta CO_2$ are compared with respective emission ratios calculated from published emission inventories (Streets et al., 2003).

2 Measurement

0 2.1 Observation site

As a part of the Program of Regional Integrated Experiments of Air Quality over Pearl River Delta (PRIDE-PRD 2006) campaign, continuous measurements of EC, CO, and CO₂ concentrations were made at Guangzhou between 1 July and 31 July 2006. Figure 1a shows an emission map of EC from Guangzhou and the surrounding region (21.75–24.25° N and 112.25–114.75° E, 0.5°) for the year 2000 (Streets et al., 2003), while Fig. 1b shows the road network in the Guangzhou urban region. In Fig. 1a and b, a crossed circle represents the observation site, located on the 16th floor (~50 m above ground level (a.g.l.)) of the Guangdong Provincial Environmental Monitoring Center (GPEMC) building (23.13° N, 113.26° E). Guangzhou city is surrounded by a highway and major roads including an express highway, ring road, and Dongfeng road pass through the urban region. Dongfeng is the road closest to the observation site. Vehicular activity on the Dongfeng road is discussed in Sect. 3.

2.2 Experimental

Measurements of EC mass concentration in the fine-mode (PM_{2.5}) were made using a semi-continuous carbon aerosol analyzer (RT 3015, Sunset Laboratory Inc., US) with

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one-hour time resolution. The analysis of elemental and organic carbon (EC-OC) was based on the National Institute for Occupational Safety and Health (NIOSH) thermaloptical transmittance (TOT) protocol for pyrolysis correction (Birch and Cary, 1996; Jeong et al., 2004; Kim et al., 2006). Ambient air samples were drawn through an inlet

- ⁵ line fitted with a cyclone with a 2.5 μ m particle diameter (PM_{2.5}) sharp cut-off size at a flow rate of 8 LPM. Then the samples were passed through a carbon-impregnated filter (CIF) multi-channel parallel-plate denuder to remove volatile organic vapors (VOCs). Samples were collected on quartz filter paper for 44 min and then heated in four stages at increasing temperatures under a helium atmosphere to quantify the carbonaceous
- ¹⁰ aerosols. As the OC vaporized during temperature ramping, it was oxidized to CO_2 in the oxidizing oven. EC was oxidized to CO_2 when the temperature was stepped up to 850°C in the oven in an oxygen environment. The instrument was calibrated by auto-injection of CH_4 (5% in He) as an internal standard. The detection limit of the instrument was estimated to be $0.4 \,\mu \text{gCm}^{-3}$, determined as three times the standard ¹⁵ deviation (σ) of a filtered air measurement (dynamic blank). The measurement uncer-
- tainty of the instrument was estimated to be 5%. The detection limit and measurement uncertainty were similar to those reported by Polidori et al. (2006), Jeong et al. (2004), Kim et al. (2006), and Jung et al. (2009) for the EC-OC analyzer.

The mixing ratio of CO was measured using a non-dispersive infrared (NDIR) ab-²⁰ sorption gas analyzer (Model 48, Thermo Environmental Instruments (TEI), US) with an integration time of one minute. CO₂ concentration was measured using an NDIRbased instrument (Model LI 7000, LI-Cor, Inc., United States) with an integration time of 10 s (Kondo et al., 2006; Takegawa et al., 2006). A common inlet line (Teflon tube of internal diameter (ID) ~6mm and length ~10m) was used for both analyzers from the

²⁵ rooftop. Because of high relative humidity levels at the measurement site in summer, air samples were passed through two sets of Nafion dryers (Perma-Pure Inc., USA) before analysis to reduce the interference of water vapor. On-site calibrations of CO and CO_2 were performed using standard mixtures with 5 ppmv of CO in air and 358 ppmv and 639 ppmv of CO_2 in air. The accuracy and precession of the CO measurements

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were 1.4% and 4 ppbv (at CO=400ppbv in 1 min), while for CO₂ these were 0.2% and 0.3 ppmv (at CO₂=400ppmv in 10 s), respectively. Further details of these analyzers were reported by Takegawa et al. (2009).

Meteorological data were obtained from the nearest meteorological station (ZDQ-13), manufactured and operated by Sun Yet-san University, China. The accuracies of meteorological parameters like wind speed, wind direction, ambient temperature, ambient pressure, and rainfall were about ±0.3ms⁻¹, ±5° (at wind speed 0.5 ms⁻¹), 0.2°C, 0.2 hPa, and 0.4 mmh⁻¹, respectively. The Mixed Layer Height (MLH) or the depth of the Planetary Boundary Layer (PBL) was estimated from the vertical profiles
of aerosol extinction coefficient at a wavelength of 1064 nm from a Light Detection and Ranging (LIDAR) system operated by the National Institute for Environmental Studies (NIES), Japan. The MLH is defined as the height where the gradient of the attenuated backscattering coefficient (ATBC) at 1064 nm is a minimum. Details of the LIDAR measurements during the PRD campaign were discussed by Sugimoto et al. (2009).

15 3 Emissions of EC, CO, and CO₂

Figure 1a shows the spatial distribution of emissions of EC. Although not shown emissions of CO and CO₂ exhibit similar distributions in Guangzhou and the surrounding urban region (Streets et al., 2003). The observation site is located within the grid (0.5°×0.5°) of highest emissions of EC, while other major emission grids are distributed
in the southeast (SE) corridors. These major EC emissions are mainly from the urban areas of Guangzhou, Foshan, Shenzhen, Zhongshan, and Jiangmen, in decreasing order. Apart from the emission sources in Guangzhou, the impact of anthropogenic activities in Foshan city could be significant, as it is the urban region closest to the measurement site. Foshan is the third largest manufacturing base in the PRD region, after Shenzhen and Guangzhou. Emissions from the sources located along the north-

west (NW) and northeast (NE) sectors are less, compared to the contributions from the southern regions. Table 1 illustrates the emissions of EC, CO, and CO_2 from domestic



and non-domestic sectors for the year 2000 (Streets et al., 2003). The domestic sector is further sub-categorized as biofuels and fossil fuels, collectively contributing about 30% of EC and 10–15% each of CO and CO₂ to the total emissions. The non-domestic sector includes emissions from industry, power generation, transport, ships, etc. and contributes about 70% of EC and 85–90% each of CO and CO. Among these sources

- ⁵ contributes about 70% of EC and 85–90% each of CO and CO₂. Among these sources emissions from industry and transport mainly contribute to the non-domestic sector (Streets et al. 2003, unpublished data). However, these inventories contain large uncertainties, viz., 484% for EC, 156% for CO, and 16% for CO₂ for China (Streets et al., 2003).
- Traffic exhaust is an important source of anthropogenic emissions in the Guangzhou urban regions (Zhou et al., 2007; Shao, 2001). Figure 2 illustrates the hourly averaged diurnal variations of traffic volume comprising heavy-duty (HDV), medium-duty (MDV), and light-duty vehicles (LDV) on the Dongfeng Middle Road recorded during 18–24 July 1999 and 12–18 October 1999 (Xie et al., 2003). These traffic data were recorded
- 7 years before the present study; however, we assume that the diurnal pattern remains nearly unchanged. The total traffic volume is lowest during early morning hours, increases after about 07:00 LT, peaks between 07:00–11:00 LT, and then decreases gradually until midnight. However, HDVs show a different pattern remaining nearly stable during the day and late evening hours (Fig. 2). There was no traffic regulation in
- ²⁰ Guangzhou for entry restrictions for heavy-duty vehicles in city area before July 2007 (http://www.chinacourt.org/flwk/show1.php?file-id=118290) as it is in Beijing to avoid the traffic jam (Han et al., 2009). It is important to note that the emission factor of EC in HDV exhaust is significantly higher than LDV and MDV (Westerdahl et al., 2009 and references therein).

²⁵ 4 Temporal variations of EC, CO, CO₂, and meteorological conditions

Figure 3 shows temporal variations of hourly averaged EC, CO, CO_2 , and some meteorological parameters. The average \pm SD of wind speed, temperature, relative humidity,

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and pressure were 2.6 ± 1 ms⁻¹, 31 ± 3 °C, $76\pm14\%$, and 1003 ± 5 hPa, respectively, during the campaign. Meteorological conditions were fairly stable, with steady wind flow (~3ms⁻¹) from the South China Sea during 2–11 July. The concentration of EC remained constant at ~4 μ gCm⁻³ for this period, except for two episodes of elevated levels in the early morning of 3 July and evening of 7 July. Observations during 12–13 July were impacted by rather weak northerly flow (<2ms⁻¹). These stagnant weather conditions could have favored the accumulation of recently emitted pollutants, resulting in higher EC and CO of ~12.5 μ gCm⁻³ and ~1700ppbv, respectively. A typhoon (named Billis) originating over the southern oceanic region hit the PRD on 15 July and

- continued until 17 July. Significant amounts of these species were likely scavenged by the rain associated with the strong wind flow during the typhoon circulation (Fig. 3). Relatively lower concentrations of pollutants were observed until 18 July due to dilution caused by the mixing of cleaner air. No significant changes in the concentrations of EC or CO were observed during 19–22 July. Later, during 23–26 July, observations
 were influenced by stagnant weather, and consequently EC and CO reached levels of 15.0 μgCm⁻³ and 2500 ppbv, respectively, on some occasions. Another typhoon
- event, named Kaemi, arrived over the PRD with strong winds and rain during 27–28 July. The long-term trend of CO₂ mixing ratio was similar to that of EC and CO. To avoid any bias, measurements conducted during the episodes of rainfall have been excluded from the statistical and correlation analyses.

Generally, the concentrations of EC, CO, and CO_2 appear to have been influenced by changes in meteorological conditions. To investigate the impact of long-range transport of the air masses, 3 – day isobaric back trajectories were calculated using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT-4) model (Draxler and

Rolph, 2003) (see Fig. 4). Each trajectory starts at 00:00 LT (local time) and 50 ma.g.l. at the measurement site. Based on the analyses of the entire period of observations, the air masses at the Guangzhou site have been classified into two main categories depending on the direction of transport, from northerly (red line in Fig. 4) and southerly (blue line in Fig. 4) directions. The southerly air masses originated over the South



China Sea (SCS) and prevailed during 1–11 July, 16–22 July, and 27–31 July, influencing ~75% of the entire campaign. Air masses categorized as northerly air (~25% of the measurements) arrived from the NW and NE directions and influenced the observations during 12–14 July and 23–26 July, which were also the periods of elevated ⁵ levels of pollutants.

Data measured in both northerly and southerly air masses are further classified into two categories, day (08:00–18:00 LT) and night (20:00–06:00 LT). The daytime measurements represent observations of well-mixed air due to both higher boundary layer depth and wind speed. Statistics of EC, CO, CO₂, and wind speed mea-¹⁰ sured in the different categories of air masses are presented in Table 2. The average ±SD (day+night) mass concentration of EC and mixing ratio of CO in the northerly air masses were $6.3\pm2.4\mu$ gCm⁻³ and 1059 ± 589 ppbv, while these were $4.3\pm1.9\mu$ gCm⁻³ and 693 ± 327 ppbv in the southerly air masses, respectively. The levels of EC and CO were about 30% higher in the northerly air masses than those in the southerly air masses. Based on all measured data the average concentrations of EC, CO, and CO₂ were $4.7\pm2.3\mu$ gCm⁻³, 798 ± 459 ppbv, and 400 ± 13 ppmv, respectively.

During northerly flow, the meteorological conditions favored stagnation, causing higher levels of pollutants in ambient air. In contrast, southerly air masses were generally dominated by cleaner marine air from the South China Sea (Liu et al., 2008; Lai

et al., 2007; Yeung et al., 2006). We have estimated the background concentrations, illustrated in Table 3. The background concentration is defined as the 1.25 percentile of the dataset (Kondo et al., 2006). The background concentrations of both EC and CO in the northerly air masses were higher by about a factor of two than those estimated for southerly air masses.

25 5 Diurnal variations

Diurnal plots of the hourly average \pm SD of EC, CO, CO₂, wind speed, and MLH for both northerly and southerly air masses are shown in Fig. 5. The diurnal variations in

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these species are particularly strong during northerly flow. The concentrations of EC, CO, and CO₂ start increasing from the early morning hours (05:00-06:00 LT), reaching their peaks at around 07:00-09:00 LT. As the day advances, the levels of these species decrease gradually during the afternoon (13:00-15:00 LT) due to higher vertical mixing

- ⁵ caused by a simultaneous increase in wind speed and MLH. In the evening hours, concentrations of EC, CO, and CO₂ exhibit increasing trends before their peaks at around 19:00–21:00 LT. Their peak concentrations coincide with the rush hours and therefore can be attributed mainly to traffic emissions. Similar features have been reported from measurements in other urban regions (Glasius et al., 2006; Park et al.,
- 2005; Latha et al., 2004; Baumgardner et al., 2007; Dutkiewicz et al., 2009). Variations of EC concentration closely follow the traffic pattern of HDVs, suggesting their major influence (Fig. 6). As we have discussed the emission factor of EC from HDVs is higher than other vehicles. Thus peaks in EC can be attributed mainly to the emissions from HDVs, as the combustion of diesel fuel in these vehicles is an important source of EC (Band et al., 2004). Emissions from ether sources mainly from industrial and demostian.
- (Bond et al., 2004). Emissions from other sources mainly from industrial and domestic activities can also contribute to the observed levels of these species (see Table 1). As can be seen in Fig. 5, EC and CO exhibit elevated concentrations and strong diurnal variations in northerly air compared to that observed during southerly flow. These characteristic differences in the features of diurnal variability could be attributed mainly
- to the variations in the regional-scale transport and meteorology. The elevated levels of pollutants in northerly air can be due to both local emissions and transport of polluted continental air from northern China. On the other hand, southerly air brings cleaner marine air, which can dilute the locally emitted pollutants, resulting in the observed lower concentrations of trace species. During the afternoon hours the MLH and wind speed
- reached maximum values; therefore, levels of these species were observed to be the lowest. The diurnal features in the two different air mass types are also supported by different meteorological conditions; for example, in the early morning hours (06:00–09:00 LT) the MLH for northerly air was shallower by ~500m than that during southerly flow. In other words, observed lower levels of EC, CO, and CO₂ in the southerly air flow

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could be due to the availability of more space for dilution. The significant differences in MLH for the two different air mass types coincide with the rush hours, and as a result the amplitudes of peaks in EC, CO, and CO_2 were much larger in northerly air. We observed that day-to-night ratios of all trace species were higher in the southerly air com-

⁵ pared to that during northerly flow. The transport from strong daytime emission sources situated in the south of Guangzhou could have counterbalanced the dilution due to increased MLH (http://www.idsgroup.com/profile/pdf/industry_series/LFIndustrial2.pdf).

6 Dependence of EC on wind and MLH

Apart from the local time dependence of the emissions, variations in meteorological parameters can influence the concentrations of pollutants; however, their contributions cannot be separated in a strict sense. Considering the significant variability in meteorology during daytime the data observed between 09:00 to 21:00 LT has been used to study the dependencies of EC on wind and MLH (Fig. 7). In the lower wind speed regime, for example from 0.5 to $1.0 \,\mathrm{ms}^{-1}$, the concentration of EC decreases sharply from about $9 \mu \text{gCm}^{-3}$ to $6 \mu \text{gCm}^{-3}$ in southerly air masses, while in the higher 15 wind speed regime, say 1.0–5.0 ms⁻¹, EC decreases gradually from $6 \mu \text{gCm}^{-3}$ to $3 \mu \text{gCm}^{-3}$. Similarly EC also decreases with wind speed in northerly air, but its level was always higher than that of southerly. Similar relationships between EC and wind speed have been observed at other urban locations, including Tokyo (Kondo et al., 2006) and Beijing (Han et al., 2009). However, the concentration of EC does not show 20 any systematic dependence on MLH for either air mass types. This relationship with MLH also explains that the levels of EC could have been influenced by strong daytime emissions. The presence of stable aerosol layers above the boundary layer could have



also inhibited the dilution of EC due to increasing MLH (Sugimoto et al., 2009).

7 Relationship between EC, CO, and CO₂

The roles of major factors impacting the diurnal variation of EC can be further investigated by estimating emission ratios of EC to other combustion products like CO and CO₂, as reported by Kondo et al. (2006) and Han et al. (2009). A diurnal plot of the slope of Δ EC/ Δ CO, calculated from a bi-variable linear fit regression method, is shown in Fig. 8. Hourly data were used for southerly air masses, while a 3-hourly interval was used for northerly air masses due to limited data. Data points with poor correlations ($r^2 < 0.35$) are not shown in Fig. 8. In the southerly air masses, the slope of Δ EC/ Δ CO increases from about 06:00 LT before a peak value (~0.011 µgCm⁻³/ppbv)

- at about 09:00 LT; later it decreases sharply and remains at about 0.005 μ gCm⁻³/ppbv during the afternoon. In the evening hours, Δ EC/ Δ CO again increases and peaks (~0.0093 μ gCm⁻³/ppbv) at 19:00 LT. The diurnal variation of Δ EC/ Δ CO closely follows the patterns of HDV and MDV traffic (see Fig. 2). Therefore the observed slope of Δ EC/ Δ CO represents the emission ratios from HDVs and MDVs particularly during
- 15 the peak hours.

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In addition to the characterization of major sources of EC, CO, and CO₂ emissions, the slopes of $\Delta EC/\Delta CO$, $\Delta EC/\Delta CO_2$, and $\Delta CO/\Delta CO_2$ can be useful in investigating the transport of EC from source regions. For example, $\Delta CO/\Delta CO_2$ ratios have been used to identify the origin of air masses (Takegawa et al., 2004). Estimates of the transport efficiency of EC from source region to the boundary layer and from the boundary layer to the free troposphere have been based on EC/CO corre-

lation slopes and emission ratios calculated from emission inventories (Sahu et al., 2008). Figure 9 shows scatter plots of the pairs EC-CO, EC-CO₂, and CO-CO₂ using all data. Table 4 illustrates details of linear fit regression slopes of Δ EC/ Δ CO, Δ EC/ Δ CO₂, and Δ CO/ Δ CO₂. The slopes of Δ EC/ Δ CO and Δ EC/ Δ CO₂ of day-time data are lower than those at nighttime in both types of air masses, whereas Δ CO/ Δ CO₂ shows the opposite pattern (Table 4). Reliable Δ EC/ Δ CO slopes are useful for assessing long-range transport of EC with reference to CO. The linear fit re-

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emissions from heavy duty-vehicles.

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8.2 Previous observations

For the first time we report hourly time-resolved measurements of EC at Guangzhou. Here we compare mean EC concentration observed in the present study with previous measurements conducted at different sites in Guangzhou during the summer (Fig. 11). The EC concentration of $4.7 \,\mu \text{gCm}^{-3}$ in this study agrees well with a range of $4.6-5.7 \,\mu \text{gCm}^{-3}$ measured at Zhongshan University (8 km from GPEMC) (Cao et al., 2003), Liwan (5 km), and Wushan (19 km) (Duan et al., 2007). Slightly higher EC concentrations of 7.9 and $6.5 \,\mu \text{gCm}^{-3}$ were observed at Huangpu (~17 km) and Long-gui (~28 km), respectively (Cao et al., 2004). Huangpu is a heavily industrialized (e.g.,

gression slopes of $\Delta EC/\Delta CO$, $\Delta EC/\Delta CO_2$, and $\Delta CO/\Delta CO_2$ over the whole dataset are 0.0054 μ gCm⁻³/ppbv, 0.15 μ gCm⁻³/ppmv, and 46.4 ppbv/ppmv, respectively.

8 Comparisons

8.1 Guangzhou and Beijing

Guangzhou and Beijing are two major urban regions of China where vehicular emissions can be important source of ambient EC. In this study, we have compared the diurnal relations of EC with traffic of heavy-duty vehicles in both cities (see Fig. 10). Similar to our observations, the diurnal variation of EC follows the traffic of heavy-duty diesel trucks (HDDTs) in Beijing (Han et al., 2009). In Beijing, the concentration of EC shows a diurnal cycle opposite to that observed in Guangzhou, as it was observed to be higher (lower) during the night (day). Similarly the traffic volume also shows the opposite diurnal patterns in these cities. Kondo et al. (2006) also report a close relationship of ambient EC with traffic in Tokyo. The present and previous studies in urban regions suggest that the concentrations of ambient EC are mainly controlled by the

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chemical, metallurgical) district of Guangzhou where emissions from power plants and vehicular exhaust are also important (Tan et al., 2006; Bi et al., 2003). Although Longgui is considered a background site, the cause of elevated EC is unknown (Cao et al., 2004). EC measured in summer is lower than the average values of 7.1 μ gCm⁻³ in spring (Andreae et al., 2008) and 14.5 μ gCm⁻³ in winter (Cao et al., 2007). A compar-5 ison of observed EC and Δ EC/ Δ CO with that reported for other major urban regions of Asia is shown in Table 5. The EC mass concentration and slopes of Δ EC/ Δ CO in Guangzhou are comparable to those observed in Beijing (China), Gwanju and Seoul (South Korea), Hyderabad (India), and Karachi (Pakistan). These results are expected as traffic emissions are known to be the major source of EC and CO in these urban re-10 gions. However, these EC concentrations are significantly higher than that measured in Tokyo. The values of $\Delta EC/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ at Guangzhou (Table 4) agree reasonably with 0.12–0.19 µgCm⁻³/ppmv and 30.2–43.9 ppbv/ppmv, respectively, in Beijing, and the slopes of $\Delta CO/\Delta CO_2$ in Guangzhou and Beijing are significantly higher than that of 11.2 ppbv/ppmv in Tokyo. This comparison suggests that measurements 15

in Tokyo are influenced by emissions from efficient fuel combustion processes.

8.3 Correlation slopes and emission ratios

To understand the importance of major emission sources impacting the levels of EC, CO, and CO₂ in the Guangzhou urban region, the slopes of Δ EC/ Δ CO, Δ EC/ Δ CO₂, and Δ CO/ Δ CO₂ are compared with the emission ratios derived from emission factors reported for different sources (Table 6) and emission inventories (Table 7). In Table 6 ranges of emission ratios of EC-CO, EC-CO₂, and CO-CO₂ are presented for different sectors. The observed slopes of Δ EC/ Δ CO (0.0054 μ gCm⁻³/ppbv), Δ EC/ Δ CO₂ (0.15 μ gCm⁻³/ppmv), and Δ CO/ Δ CO₂ (46.4 ppbv/ppmv) in this study at Guangzhou are within the range of emissions ratios estimated for diesel fuel and gasoline combustion in the transport sector. However, contributions from combustion processes in other



sectors can also influence the observed slopes of these species.

Comparison of observed slopes of $\Delta EC/\Delta CO$, $\Delta EC/\Delta CO_2$, and $\Delta CO/\Delta CO_2$ with their emission ratios derived from the inventories can be useful to validate the inventory data. However, this study has its limitations because ambient air may not be homogenously mixed as various emissions sources are collocated. The emission ratios of EC/CO, EC/CO₂, and CO/CO₂ derived for the urban regions of Guangzhou 5 (21.75-24.25° N, 112.25-114.75° E), Beijing, and Tokyo using Streets et al. (2003) inventory data for the year 2000 are shown in Table 7. These emission ratios were derived by summing the emissions from the domestic and non-domestic sectors (Streets et al., 2003). For Guangzhou, the observed slopes of $\Delta EC/\Delta CO$, $\Delta EC/\Delta CO_2$, and $\Delta CO/\Delta CO_2$ agree reasonably with the respective emission ratios (Tables 4 and 7). 10 Therefore, the observed slopes validate the emissions inventories of EC, CO, and CO₂, to some extent. However, this statement may not be entirely conclusive considering the limited measurements for this study and uncertainties in the inventories. In support of our discussion, the estimated EC/CO emissions ratio of $0.0049 \mu \text{gCm}^{-3}/\text{ppbv}$ from the transport sector (Streets et al., 2003; unpublished dataset) agrees well with the ob-15 served slope of 0.0054 μ gCm⁻³/ppbv. Emission ratios of the Guangzhou urban region

also agree well with those estimated for Beijing but not with those of Tokyo (Table 7).

9 Conclusions

As part of the PRIDE-PRD 2006 campaign, measurements of EC mass concentration, ²⁰ mixing ratios of CO and CO₂, and meteorological parameters were conducted at an urban site in Guangzhou, China during July 2006. Guangzhou is the main industrial and commercial city of the PRD region, where emissions due to road transport, industrial, and domestic activities are the major sources of pollutants. The traffic volume was highest during morning hours; however, unlike other categories of vehicles, traffic ²⁵ of heavy-duty vehicles was remained high during the daytime. The average concentra-

tions of EC, CO, and CO₂ were 4.7 μ gCm⁻³, 798 ppbv and 400 ppmv, respectively, during the campaign. Trends in EC, CO, and CO₂ concentrations were influenced by local



meteorology and episodes of typhoons. The trajectory analysis suggests the influence of cleaner marine air from the southern direction during most of the campaign, while stagnant conditions prevailed during northerly flow leading to observed higher background levels of EC and CO by a factor of about two than those during southerly flow.

- ⁵ The diurnal patterns of EC, CO, and CO_2 exhibited primary peaks during the morning and secondary peaks in the evening hours coinciding with rush-hour traffic. In addition to the traffic emissions the variability in meteorological parameters played a key role in the observed diurnal variations of these species. The diurnal patterns of EC and $\Delta EC/\Delta CO$ closely followed the patterns of HDV and MDV traffic. Similar to studies re-
- ¹⁰ ported for Beijing and Tokyo cities, EC showed a close relationship with the heavy-duty vehicle traffic in Guangzhou. The EC measured in the present study agrees reasonably well with previous measurements reported for summer at nearby sites in Guangzhou. Similar values of EC and slopes of $\Delta EC/\Delta CO$ are comparable to those reported at several urban regions in Asia. Observed slopes of $\Delta EC/\Delta CO$ (0.0054 μ gCm⁻³/ppbv), $\Delta EC/\Delta CO_2$ (0.15 μ gCm⁻³/ppmv) and $\Delta CO/\Delta CO_2$ (46.4 ppbv/ppmv) agree reason-
- ably with respective emission ratios, validating the inventories to some extent.

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Table 1. Relative contributions (in %) of EC, CO, and CO₂ emissions from different sources in Guangzhou urban area (21.75-24.25 ° N and 112.25 - 114.75° E) for the year 2000 (Streets et al., 2003)^a.

	Domes	stic sector ^a		Non-domestic sector ^b				
	Fossils fuel	Biofuel	Industry	Power	Transportation	Total		
EC	10 (2.0)	21 (4.1)	38 (7.8)	2 (0.5)	28 (5.8)	100 (20.4)		
CO	3 (101.7)	10 (337.8)	47 (1624.1)	2 (66.1)	39 (1347.4)	100 (3477.2)		
CO ₂	5 (8140.0)	6 (10 020.6)	32 (50 481.0)	39 (61 507.4)	17 (26 540.1)	100 (156 689.1)		

() Emissions of each species (in Ggyr⁻¹) ^a Streets et al. (2003) (published data)

^b Streets et al. (2003) (unpublished inventory dataset)

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Table 2. Statistics of EC, CO, CO_2 , and wind speed, observed in different categories at Guangzhou.

		EC (µgCm ⁻³) CO (ppbv)						CO ₂ (ppmv)				Wind speed (ms ⁻¹)					
		Min.	Max.	Mean±SD	Median	Min.	Max.	Mean±SD	Median	Min.	Max.	Mean±SD	Median	Min.	Max.	Mean±SD	Median
North	Day	2.7	12.9	5.8±2.4	5.2	429	2282	1025±441	909	383	452	402±14	400	0.9	4.8	2.9±0.9	2.8
	Night	3.9	18.7	6.8±2.5	6.3	443	3107	1092±737	803	383	446	410±14	408	0.4	5.7	2.4±0.9	2.4
South	Day	1.2	9.7	4.1±1.4	3.8	315	2729	734±315	688	373	436	396±11	395	0.6	5.1	2.7±0.9	2.5
	Night	1.1	17.0	4.4±2.4	3.8	263	1991	652±338	556	380	452	397±10	395	0.2	4.0	2.2±0.9	2.3
All data	a	1.1	18.7	4.7±2.3	4.2	202	3162	798±459	685	373	453	400±13	398	0.2	5.7	2.6±1.0	2.5

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Table 3. The background concentrations (1.25 percentiles) of EC, CO, and CO₂.

		EC (μ gCm ⁻³)	CO (ppbv)	CO ₂ (ppmv)
North	Day	2.79	430	383
	Night	3.88	448	386
South	Day	2.33	333	376
	Night	1.57	295	383
All data	L	1.92	322	379

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Table 4. Observed EC-CO, EC-CO₂, and CO-CO₂ linear regressions slopes (\pm 1SD).

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		$(\mu g Cm^{-3}/ppbv)$	r^2	$(\mu g Cm^{-3}/ppmv)$	r^2	(ppbv/ppmv)	r^2
North	Day	0.0045±0.0004	0.68	0.13±0.1	0.62	49.4±3.1	0.60
	Night	0.0069±0.0008	0.58	NA	—	NA	—
South	Day	0.0051 ± 0.0005	0.59	0.12±0.1	0.55	57.4±1.9	0.46
	Night	0.0070 ± 0.0004	0.75	0.21±0.1	0.76	34.9±1.5	0.67
All data	Day	0.0045 ± 0.0005	0.51	0.13±0.1	0.57	53.2±1.9	0.50
	Night	0.0067 ± 0.0004	0.72	0.16±0.1	0.55	NA	—
All data		0.0054±0.0002	0.61	0.15±0.1	0.58	46.4±1.5	0.45

NA: not available

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Table 5. The concentrations of EC aerosol and linear fit regression slopes measured in major urban regions of Asia.

Place and measurement period	EC (μgCm ⁻³)	$\Delta EC/\Delta CO$ (µgCm ⁻³ /ppbv)	Method of EC analysis	References
Guangzhou, China (Jul 2006)	4.7	0.0054	TOT	Present study
Guangzhou, China (Oct–Nov 2004)	7.1	0.0079	TOT	Andreae et al. (2008)
Beijing, China (2005–2006)	6.9	0.0035-0.0058	TOT	Han et al. (2009)
Tokyo, Japan (2003–2005)	1.9	0.0057	TOT	Kondo et al. (2006)
Gwanju, South Korea (Mar-May 2001)	5.7	0.0060	TOT	Park et al. (2005)
Seoul, South Korea (1998–1999)	4.5	0.0022	TMO	Park et al. (2007)
Hyderabad, India (Jan 2004)	1.5–11.2	0.0073	L. Abs.	Latha et al. (2004)
Karachi, Pakistan (2006–2007)	5.9	NA	L. Abs.	Dutkiewicz et al. (2009)

(): *r*²

NA: not available TOT: thermal-optical-transmittance TMO: Thermal manganese dioxide oxidation L. Abs.: Light absorption **Table 6.** EC/CO (μ gCm⁻³/ppbv), EC/CO₂ (μ gCm⁻³/ppmv), and CO/CO₂ (ppbv/ppmv) emissions ratios derived from emission factors (grams of pollutant evolved per kilogram of fuel burned).

	Trar	nsport	Industry	Dom	estic	Biomass burning
	Diesel ^{d,e,g,h}	Gasoline ^{d,h}	Coal ^d	Coal ^{d,f}	biofuels ^{c,d}	(crop residue) ^{a,b,c}
EC/CO	0.0013-0.055	0.0031-0.0115	0.0017-0.0182	0.0019-0.0572	0.0087-0.0266	0.0056-0.016
EC/CO ₂	0.15	0.06-0.11	0.04-0.74	0.11-3.56	0.69-1.75	0.53-1.10
CO/CO_2	8.6-65.2	33.5	23.5-40.4	53.3-62.2	52.9–98.5	45.7-123.6

^a Dhammapala et al. (2007)

^b Cao et al. (2008)

^c Andreae and Merlet (2001)

^d Streets et al. (2003)

^e Dickerson et al. (2002)

^f Boucher et al. (2008)

^g Sanchez et al. (2009)

^h Westerdahl et al. (2009)

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Table 7. Emissions ratios derived from the inventory of Streets et al. (2003) for major urban regions in Asia.

	EC/CO (µgCm ⁻³ /ppbv)	EC/CO ₂ (μ gCm ⁻³ /ppmv)	CO/CO ₂ (ppbv/ppmv)
Guangzhou	0.0067	0.23	35.0
Beijing	0.0050	0.24	47.6
Tokyo	0.0093	0.082	8.8



Fig. 1. (a) Spatial distribution of EC emissions (tons/yr) in Guangzhou and surrounding areas (21.75–24.25° N and 112.25–114.75° E). Numeric labels are for: 1. Guangzhou, 2. Foshan, 3. Zhouqing, 4. Qianguan, 5. Huizhou, 6. Dongguan, 7. Shenzhen, 8. Zhuhai, 9. Zhongshan, 10. Jiangmen. White circled cross represents observation site. **(b)** Road network surrounding the observation site in Guangzhou.

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Fig. 2. Diurnal patterns of heavy-duty (HDV), medium-duty (MDV) and light-duty (LDV) vehicles on the Dongfeng middle road, recorded during 18–24 July 1999 and 12–18 October 1999 (Xie et al., 2003).

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Fig. 5. Diurnal variations of hourly average \pm SD of EC, CO, CO₂, wind speed, and mixed layer height (MLH).





Fig. 6. Diurnal variations of hourly EC and heavy-duty vehicles (HDV) in Guangzhou.

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Fig. 7. Dependencies of EC on wind speed and MLH, calculated for each 0.5 ms^{-1} of wind speed and 100 m (200 m for north) bins of MLH, respectively. Data observed between 09:00 to 21:00 LT have been used for the calculation of dependencies of EC.

0.014 -1.0 - North ΔEC/ΔCO (μgC m⁻³/ppbv) Correlation Coefficient (r² 0.012 ► South -0.9 0.010 -0.8 0.008 -0.7 0.006 -0.6 0.004 0.5 0.002 -0.4 0.000 00:00 06:00 12:00 18:00 00:00 Local time

Fig. 8. Diurnal variations of $\Delta EC/\Delta CO$ linear regression slopes (±SD bar) (3-h for northern and 1-h for southern air masses) and r^2 . Missing data points are for $r^2 < 0.35$.

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