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Source areas and chemical composition of fine particulate matter in the Pearl River Delta region of China

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Abstract

Fine particulate matter ($PM_{2.5}$) was measured for 4 months during 2002–2003 at seven sites located in the rapidly developing Pearl River Delta region of China, an area encompassing the major cities of Hong Kong, Shenzhen and Guangzhou. The 4-month average fine particulate matter concentration ranged from 37 to 71 $\mu g\,m^{-3}$ in Guangdong province and from 29 to 34 $\mu g\,m^{-3}$ in Hong Kong. Main constituents of fine particulate mass were organic compounds (24–35% by mass) and sulfate (21–32%). With sampling sites strategically located to monitor the regional air shed patterns and urban areas, specific source-related fine particulate species (sulfate, organic mass, elemental carbon, potassium and lead) and daily surface winds were analyzed to estimate influential source locations. The impact of transport was investigated by categorizing 13 (of 20 total) sampling days by prevailing wind direction (southerly, northerly or low wind-speed mixed flow). The vicinity of Guangzhou is determined to be a major source area influencing regional concentrations of $PM_{2.5}$, with levels observed to increase by 18–34 $\mu g\,m^{-3}$ (accounting for 46–56% of resulting particulate levels) at sites immediately downwind of Guangzhou. The area near Guangzhou is also observed to heavily impact downwind concentrations of lead. Potassium levels, related to biomass burning, appear to be controlled by sources in the northern part of the Pearl River Delta, near rural Conghua and urban Guangzhou. Guangzhou appears to contribute 5–6 $\mu g\,m^{-3}$ of sulfate to downwind locations. Guangzhou also stands out as a significant regional source of organic mass (OM), adding 8.5–14.5 $\mu g\,m^{-3}$ to downwind concentrations. Elemental carbon is observed to be strongly influenced by local sources, with highest levels found in urban regions. In addition, it appears that sources outside of the Pearl River Delta contribute a significant fraction of overall fine particulate matter in Hong Kong and Guangdong province. This is evident in the

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relatively high $PM_{2.5}$ concentrations observed at the background sites of 18 $\mu g\,m^{-3}$ (Tap Mun, southerly flow conditions) and 27 $\mu g\,m^{-3}$ (Conghua, northerly flow conditions).

Keywords: Pearl River Delta region; $PM_{2.5}$; Chemical composition; Source regions; Transport

1. Introduction

While the rapid development of the Pearl River Delta Region (PRD) of China has elevated the living conditions of many Chinese citizens in both Hong Kong and Guangdong province, the fast-paced growth in population and energy use has been paralleled by the degradation of local air quality. One pollutant strongly impacted by growth in anthropogenic sources is fine particulate matter, $PM_{2.5}$. Atmospheric fine particulate pollution, produced primarily through combustion processes, has received the attention of governments and research programs around the world for its impact on human respiratory health (Dockery et al., 1993; Englert, 2004; Pope et al., 2002; Pozzi et al., 2003; Schwartz et al., 1996), visibility and climate (Chameides and Bergin, 2002; Chameides et al., 1999; Charlson et al., 1992; Haywood and Shine, 1995; Schwartz, 1996). With a lifetime in the lower atmosphere of days to weeks, $PM_{2.5}$ can be transported thousands of kilometers, complicating policy development to alleviate fine particle pollution in a target region.

Home to a population of 46 million, the PRD encompasses the floodplains region of the Pearl River (Zhujiang) in the southern province of Guangdong as well as the southward mountainous New Territories and island regions of Hong Kong. With Hong Kong established as a service sector (Cullinane and Cullinane, 2003) and Guangdong province focused on manufacturing and energy production (Warren-Rhodes and Koenig, 2001), the intertwined economies have proven to be mutually beneficial to all members of the delta, where the regional GDP grew at a rate of ~17% per year from 1980 to 2000 (Cullinane and Cullinane, 2003). Such phenomenal growth, however, has also yielded high concentrations of $PM_{2.5}$ in many areas of the PRD, an issue of concern to local governments and citizens.

Without well-established information on major sources and source locations in the PRD, the majority of past studies have used ground-level monitoring to study the nature of regional fine particulate pollution, with the majority of these field campaigns limited to the Hong Kong area. Measurements by a number of research groups (Cao et al., 2003, 2004; Chan et al., 2001; Chao and Wong, 2002; Ho et al., 2002, 2003; Louie et al., 2005; Wei et al., 1999) have observed fine particle concentrations at various sites in the PRD consistently exceeding the annual US National Ambient Air Quality Standard of 15 $\mu g\,m^{-3}$. A recent field campaign conducted by Cao et al. (2003, 2004) sampled for $PM_{2.5}$ and carbonaceous species (EC and OC) during the Winter and Summer of 2002 in four cities of the PRD, finding highest $PM_{2.5}$ concentrations in Guangzhou (106 $\mu g\,m^{-3}$), followed by Shenzhen (61 $\mu g\,m^{-3}$), Zhuhai (59 $\mu g\,m^{-3}$) and Hong Kong (55 $\mu g\,m^{-3}$).

Though several researchers have found evidence that sulfate concentrations in Hong Kong are influenced by regional sources (Ho et al., 2003; Louie et al., 2005; Qin et al., 1997), with more localized sources impacting the carbonaceous component (Louie et al., 2005), the location of major $PM_{2.5}$ sources in the PRD has yet to be determined. In order to guide regional air quality management, an in-depth study linking regional $PM_{2.5}$ chemical characteristics with influencing factors (sources, source locations and meteorology) is needed. This paper presents an overview of a large-scale monitoring study that took place for 4 months (1 month per season) over the time span of October 2002 to June 2003, with simultaneous measurements of $PM_{2.5}$ mass and chemical composition at seven sites in the PRD. Measurements of local meteorology are combined with the daily concentrations of source-related fine particulate species to approximate the location of influential sources and to better understand the impact of specific wind patterns on the resulting regional $PM_{2.5}$ concentrations.

2. Experiment methodology

Measurements were conducted for 1 month per season at seven sites located in Hong Kong Special Administrative Region and Guangdong province.

The sites, three in Hong Kong and four in Guangdong, were selected to represent background concentrations, urban sources and receptor areas downwind of the major urban sources. The specific sites and their characteristics are listed in Table 1 with locations shown in a regional map displayed in Fig. 1. Also shown on the regional map are meteorological stations providing our project with measurements of hourly wind speed and direction as well as 24 h precipitation.

At each selected sampling site, 24 h $PM_{2.5}$ samples intended for later analysis of mass concentration and composition were collected every sixth day (five samples per month) for the months of October 2002, December 2002, March 2003 and June 2003. The sampling sites of Tap Mun (TM), Tung Chung (TC), central and western (CW), Guangzhou (GZ) and Shenzhen (SZ) used ThermoAndersen RAAS $PM_{2.5}$ chemical speciation samplers, while Conghua (CH) and Zhongshan (ZS) used Caltech Gray Box samplers that served as the prototype design for the RAAS $PM_{2.5}$ sampler. The Caltech Gray Box prototype, which has been described previously (Chowdhury et al., 2001), removes coarse particles

Table 1
Sampling sites in the Pearl River Delta

Site	Site code	Location	Coordinates	Site description
Tap Mun	TM	Hong Kong	Lat: N 22 28.277 Lon: E 114 21.642	Background
Tung Chung	TC	Hong Kong	Lat: N 22 17.324 Lon: E 113 56.620	Downwind of urban source
Central and western	CW	Hong Kong	Lat: N 22 17.122 Lon: E 114 08.713	Urban source
Shenzhen	SZ	Guangdong	Lat: N 22 32.183 Lon: E 113 56.340	Urban source
Zhongshan	ZS	Guangdong	Lat: N 22 30.660 Lon: E 113 24.429	Downwind of urban source
Guangzhou	GZ	Guangdong	Lat: N 23 07.969 Lon: E 113 15.585	Urban source
Conghua	CH	Guangdong	Lat: N 23 38.530 Lon: E 113 38.345	Background

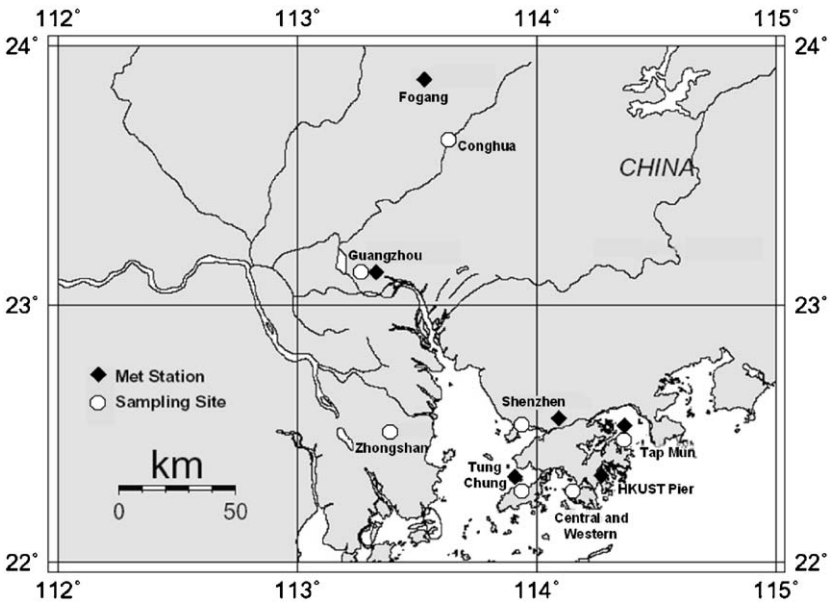

Fig. 1. Location of sites used for the PRD study.

via a cyclone separator (John and Reischl, 1980). Both the ThermoAndersen RAAS and Caltech Gray Box $PM_{2.5}$ samplers collect fine particles onto filters in four channels (two 47 mm quartz filters, two 47 mm Teflon filters) via two separate flows of $24\,L\,min^{-1}$ passing through a $PM_{2.5}$ cyclone, followed by a manifold splitting each flow line into a quartz filter channel ($16.7\,L\,min^{-1}$ for RAAS $PM_{2.5}$ and $14.0\,L\,min^{-1}$ for Caltech Gray Box) and a Teflon filter channel ($7.3\,L\,min^{-1}$ for RAAS $PM_{2.5}$ and $10.0\,L\,min^{-1}$ for Caltech Gray Box). Flow rates were controlled by critical orifices located upstream of a vacuum pump and were measured periodically throughout each sampling month with a calibrated dry gas meter (NIST Traceable-ID #C-0701).

Intercomparison sampling performed between co-located Caltech Gray Box and ThermoAndersen RAAS samplers demonstrated measurements of $PM_{2.5}$ mass concentration to be within 5% of a reference ThermoAndersen RAAS sampler and thus suitable for joint use in the PRD field measurement campaign. Operators at each of the seven sampling sites were carefully trained in filter handling and storage. After each sampling period, filter samples were sealed in Petri dishes and stored under freezing temperatures to minimize the loss of volatile species. Samples were later transported in ice-packed coolers to their eventual destinations in Hong Kong and California for mass measurement and chemical analyses. To track any contamination due to handling, four field blanks (one per sampling month) were taken at each site. The blank filters were stored and transported alongside the 24 h samples.

3. Chemical analyses

Following sample collection at each of the seven locations, the Teflon filters were analyzed for mass concentration, major ions (sulfate, nitrate, chloride, ammonium) and trace elements. Quartz filters were used to perform detailed speciation of organics as well as to measure elemental carbon and organic carbon. Teflon filter mass measurements were conducted via a microbalance (Mettler Instruments) following the guidelines of the EPA Quality Assurance Document 2.12 (EPA, 1998) and repeat measurements were performed on all samples to ensure accurate results. Major ion concentrations (SO_4^{2-}, NO_3^- and Cl^-) were determined via ion chromatography, comparing sampled concentrations with laboratory standards prepared from ACS grade analytical reagents. For the measurement of ammonium ion (NH_4^+), indophenol colorimetric analysis was performed with a rapid flow analyzer (RFA-300 TM, Alpkem Corporation) (Bolleter et al., 1961). Trace elements were determined using X-ray fluorescence (XRF) analysis (Watson et al., 1996). Quartz fiber filters were analyzed with a carbon analyzer (Sunset Laboratory) for elemental carbon (EC) and organic carbon (OC) using the established NIOSH protocol of thermal evolution and combustion, initially developed by Birch and Cary (1996). A correction factor of 1.4 is applied to measured organic carbon to estimate overall organic mass (OM). While the OM to OC ratio is not necessarily a constant value in an ambient aerosol population, the value of 1.4 has been suggested by recent research as an appropriate adjustment factor to reconcile OC values to the original mass of organic compounds (Russell, 2003).

4. Results and discussion

4.1. Consolidated averages

Consolidated averages of 20 filter samples resulted in $PM_{2.5}$ concentrations in Guangdong province ranging from $37\,\mu g\,m^{-3}$ at rural Conghua to $71\,\mu g\,m^{-3}$ at urban Guangzhou, a much smaller range is observed in Hong Kong from $29\,\mu g\,m^{-3}$ at rural Tap Mun to $34\,\mu g\,m^{-3}$ at urban Central/Western. Averages of chemical composition, presented in Table 2, show that fine particulate mass within the PRD is dominated by organic compounds (24–35%) and sulfate (21–32%). Other important measured constituents include crustal material (7–13%), ammonium (6–8%), elemental carbon (3–8%) and nitrate (1–6%).

Particulate OM, formed by both direct emissions and through gas-to-particle conversion, ranged annually from 6.9 to $9.3\,\mu g\,m^{-3}$ at sites in Hong Kong, and from 12.7 to $24.6\,\mu g\,m^{-3}$ in Guangdong. As shown in Table 2, annual mean elemental carbon (EC) concentrations were low at background sites Conghua ($1.4\,\mu g\,m^{-3}$) and Tap Mun ($0.8\,\mu g\,m^{-3}$), compared with a range of 1.9–$4.4\,\mu g\,m^{-3}$ for the five sites in more developed locations. Annual average OM/EC ratios had maximum values at background sites Conghua (9.0) and Tap Mun (8.4), with lower values of 4.0–5.9 for the remaining areas of the region. Previous studies have also measured the carbonaceous component to $PM_{2.5}$ in Hong Kong and Guangdong, although different OC and EC

Table 2
Mean fine particulate matter concentrations across the PRD

Site[b]		Mean concentration ($\mu g\,m^{-3}$)[a]																	
	N	$PM_{2.5}$		OM[c]		EC		Nitrate		Sulfate		Ammonium		Crustal[d]		Trace[e]		Other	
TM	20	28.7	(13.8)	6.9	(3.9)	0.8	(0.5)	0.5	(0.3)	9.2	(4.7)	2.3	(1.2)	2.3	(1.7)	0.9	(0.4)	5.9	(4.5)
TC	20	32.5	(14.8)	8.8	(4.7)	2.0	(1.0)	0.8	(0.8)	9.0	(5.1)	2.5	(1.3)	2.2	(1.6)	1.0	(0.4)	6.1	(5.1)
CW	20	34.3	(14.1)	9.3	(4.3)	1.9	(1.0)	1.0	(0.7)	9.3	(4.5)	2.5	(1.2)	2.4	(1.6)	1.0	(0.3)	7.0	(4.5)
SZ	21	47.1	(21.3)	15.6	(7.9)	3.9	(2.3)	2.3	(1.8)	10.0	(5.3)	3.2	(1.6)	4.1	(2.3)	1.6	(0.9)	6.6	(5.1)
ZS	19	46.5	(23.0)	14.7	(9.1)	2.5	(1.2)	1.8	(1.8)	11.9	(5.7)	3.3	(1.6)	4.5	(3.5)	1.4	(0.9)	6.4	(6.1)
GZ	20	70.6	(30.2)	24.6	(11.6)	4.4	(1.8)	4.0	(3.7)	14.7	(6.1)	4.5	(2.2)	6.6	(3.1)	2.5	(1.3)	9.3	(7.1)
CH	19	36.8	(18.5)	12.7	(6.3)	1.4	(0.5)	0.3	(0.2)	10.4	(5.4)	2.4	(1.2)	4.6	(2.3)	1.4	(0.5)	3.7	(3.6)

[a]Values in parentheses represent standard deviation.
[b]Site labels are as follows: Tap Mun (TM), Tung Chung (TC), central/western (CW), Shenzhen (SZ), Zhongshan (ZS), Guangzhou (GZ) and Conghua (CH).
[c]OM (organic mass) is calculated by multiplying the measured organic carbon by 1.4.
[d]Crustal is the sum of the common oxides of Si, Al, Fe, Ti, Mn, Ca and K, and uses a factor of 1.17 to include carbonate and the water of hydration.
[e]Trace species include elements determined from XRF analysis not included in "crustal", converted to their oxide forms where appropriate.

detection methods were used and thus the data cannot be directly compared. Measurements in Hong Kong by Louie et al. (2005) and throughout both Hong Kong and Guangdong by Cao et al. (2003, 2004) utilized thermal evolution analysis based on laser reflectance (IMPROVE) with a different temperature program than the laser transmittance method (NIOSH) that is used in this study. Chow et al. (2001) verified experimentally that the two methods have similar results for total carbon but variability exists in the division of carbon between EC and OC. Ho et al. (2003) measured EC and OC in Hong Kong using thermal manganese dioxide oxidation (TMO method), which has been found to closely compare to results from the IMPROVE protocol (Fung et al., 2002).

Average particulate sulfate concentrations, generated primarily through the oxidation of emitted gaseous SO_2, had lower values and a smaller range in Hong Kong (9.0–9.3 $\mu g\,m^{-3}$) compared with that in Guangdong (10.0–14.7 $\mu g\,m^{-3}$). The close range in measured sulfate at sites representing both urban and background locations in Hong Kong supports the notion that sulfate has regional and perhaps longer range sources. Our measurements at sites in Hong Kong are similar to those by Louie et al. (2005), who measured average $PM_{2.5}$ sulfate concentrations at three locations in Hong Kong in the range of 8.7–9.4 $\mu g\,m^{-3}$ during 2000–2001. In comparing sulfate and ammonium ion concentrations, it is observed that the 4-month average ammonium/sulfate molar charge ratio ranges from 0.60 to 0.85 among the seven sites in the PRD. This indicates that the aerosol phase is acidic, with ammonium principally associated with sulfate. A recent emissions inventory by Streets et al. (2003) found agricultural activities to be the main source of ammonia in China, which is the likely explanation for the higher levels of ammonium at the sites in Guangdong compared with Hong Kong.

With overall average concentrations of crustal and trace species shown in Table 2, two specific species related to anthropogenic activities, lead (fuel combustion, industrial sources) and potassium (biomass burning), will be discussed in further detail. While fine particulate potassium can also be derived from crustal material, the high portion of water soluble potassium (~90%) in Hong Kong's $PM_{2.5}$ reported by Louie et al. (2005) indicates that crustal sources have a limited influence on resulting potassium levels. Lead and potassium are both at trace levels in samples, though are frequently linked with other particulate species (OM, EC) that constitute a larger portion of fine particulate mass. It is of note that consolidated averages of lead in the PRD have lower levels in Hong Kong (0.05–0.06 $\mu g\,m^{-3}$) compared with Guangdong (0.08–0.26 $\mu g\,m^{-3}$). Lead concentrations in Hong Kong are similar to past measurements by Louie et al. (2005), who found a range of 0.06–0.07 $\mu g\,m^{-3}$ at three sites in Hong Kong during year-long monitoring in 2000–2001. Overall lead concentrations in the PRD are similar to elsewhere in

Asia, with wintertime levels at a background site in Korea measuring 0.03 and 0.20 $\mu g\,m^{-3}$ observed at urban Seoul (Mishra et al., 2004). He et al. (2004) reported high average lead concentrations in Beijing, with an annual average at a residential area in the city (0.33 $\mu g\,m^{-3}$) exceeding our consolidated average at Guangzhou (0.26 $\mu g\,m^{-3}$). As with lead, potassium concentrations in Hong Kong (0.56–0.57 $\mu g\,m^{-3}$) are lower than that measured northward in Guangdong (0.74–1.60 $\mu g\,m^{-3}$). The presented potassium values in Hong Kong are comparable with the range of 0.49–0.58 $\mu g\,m^{-3}$ measured during 2000–2001 by Louie et al. (2005). Linked with biomass burning, measured potassium concentrations in the PRD are within the range of values measured during biomass burning events in Korea (0.49 $\mu g\,m^{-3}$ for the burning of rice straw, 4.19 $\mu g\,m^{-3}$ for burning of barley) and more than double levels observed during non-burning periods (0.25 $\mu g\,m^{-3}$) (Ryu et al., 2004).

4.2. Meteorology case studies

In order to provide insight into source area locations as well as meteorological influences (i.e. wind speed and precipitation) on fine particulate concentrations, daily surface wind patterns are compared with concentrations of fine particulate species at the seven sites in the PRD. In the four months of sampling, three unique meteorological cases were identified which can be summarized as follows: (1) "southerly flow" characterized by low to moderate winds from the South; (2) "northerly flow" having moderate to strong winds from the North, and (3) "mixed flow" associated with weak winds (wind speed $<3\,m\,s^{-1}$) shifting in direction throughout the day. Table 3 lists the 13 sampling days (of 20 total) categorized into each meteorological case, with the remaining 7 days excluded for not clearly fitting into one of the three identified categories or for inconsistency in wind measurements among monitoring sites. Measured hourly wind speed, wind direction and daily precipitation for each categorized day are shown in Fig. 2. Though some variability exists among the meteorology sites, the presented measurements at the Shenzhen meteorology station (Lat: 22.5500, Long: 114.1000) represent general observed trends in wind and precipitation at the five other meteorological sites shown in Fig. 1. It should be noted that surface wind measurements do not necessarily represent large-scale flow patterns, as local topography can affect surface measurements. However, the selected meteorological sites in the PRD are strategically placed to represent regional rather than local winds.

Table 3
Sampling days categorized into southerly, northerly or mixed flow

Meteorological cases		
Southerly flow	Northerly flow	Mixed flow
2002.10.20	2002.10.08	2002.10.14
2003.06.05	2002.12.13	2002.12.19
2003.06.23	2002.12.25	2003.03.13
2003.06.29	2003.03.07	2003.03.25
		2003.06.17

To relate upwind source regions with downwind concentrations of fine particulate matter, specific species linked with sources are examined for each meteorological category, including sulfate (coal combustion), organic compounds (combustion of fossil fuels, biomass burning, industrial sources, local cooking), elemental carbon (poor coal combustion, fuel oil combustion, combustion of diesel gasoline), potassium (biomass burning) and lead (combustion of leaded gasoline, industrial sources). In order to compare the grouped series of days, each daily measured species is normalized by the concentration of the identical species on the same day at the Guangzhou site and then the relative concentrations of the grouped days are averaged. Guangzhou was selected as a reference site because it is centrally located, has the highest average fine particulate matter concentration among the seven monitoring sites and is hypothesized to be a major source area contributing to downwind concentrations. The normalization lessens any bias due to precipitation events and seasonally changing source strengths, such as the variability of biomass burning events throughout the year. Assuming that source locations are remaining constant, the normalization allows a clear view into impacts of wind patterns on relative concentrations among the seven sites. Shown in Fig. 3, the average relative concentration and standard error of the selected species at each site are categorized into southerly, northerly and mixed flows. Also, non-normalized average concentrations for each meteorological case are shown in Table 4, though it should be noted that looking at a single site's change between meteorological cases may be biased by an uneven distribution of precipitation events within each category.

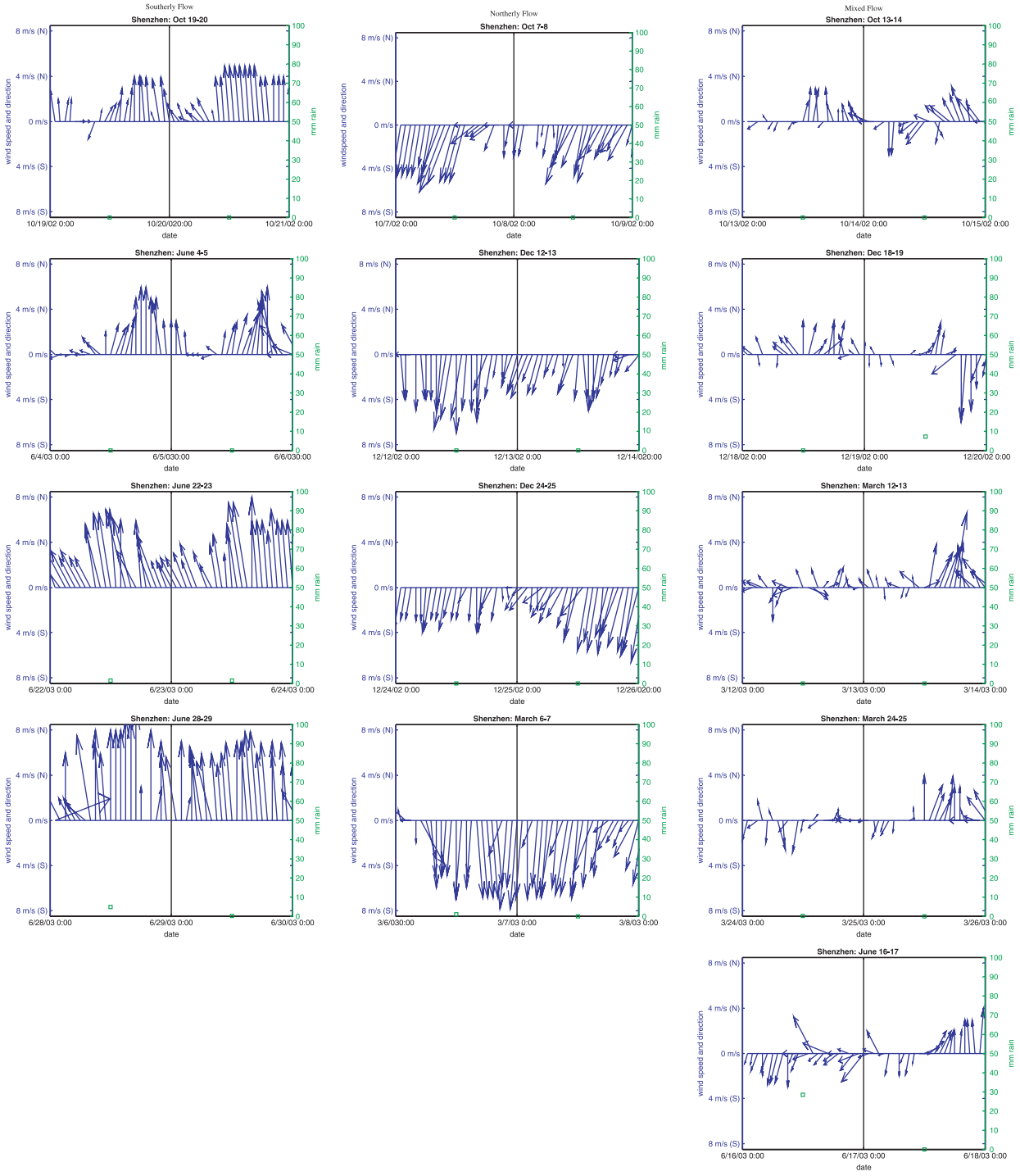

Fig. 2. Measurements of wind speed, wind direction and precipitation at the Shenzhen meteorology site for selected days categorized into northerly, southerly and mixed flow. Hourly wind magnitude and direction are indicated by the arrows and 24 h rainfall is indicated by the square markers.

4.2.1. *Southerly flow*

Relative concentrations of $PM_{2.5}$ and specific chemical species for southerly flow conditions can be seen in the leftmost column of graphs in Fig. 3. Average values for each of the flow conditions are given in Table 4. During days with southerly winds

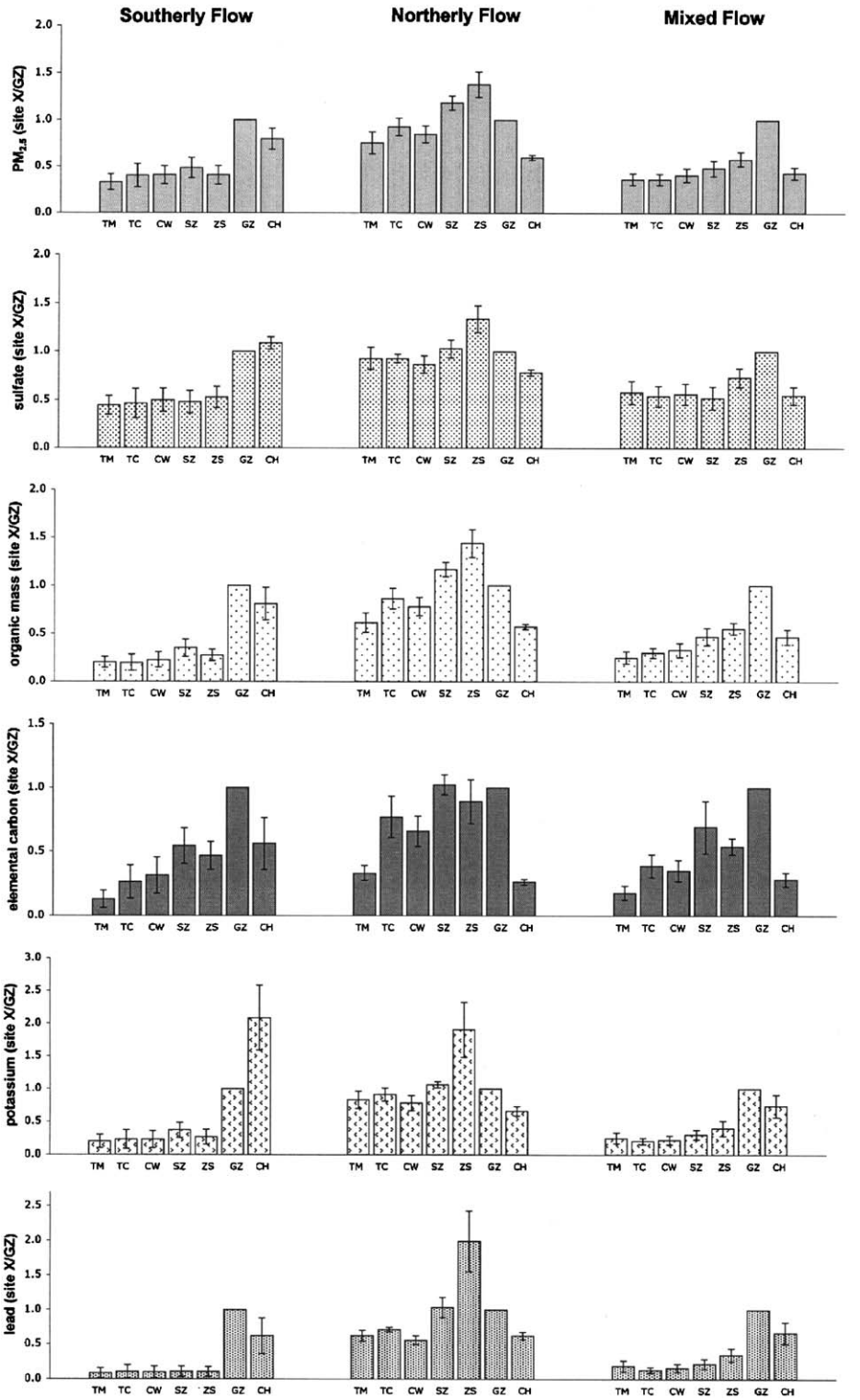

Fig. 3. Normalized concentrations and standard error of species measured at seven sites in the Pearl River Delta, categorized by wind pattern. Site labels are as follows: Tap Mun (TM), Tung Chung (TC), central/western (CW), Shenzhen (SZ), Zhongshan (ZS). Guangzhou (GZ) and Conghua (CH).

Table 4
Average concentrations of measured species during southerly, northerly and mixed flow

Site	Average concentration in mg m^{-3}					
	$PM_{2.5}$	Sulfate	OM	EC	Potassium	Lead
Southerly flow						
Tap Mun	18.0	6.0	3.3	0.30	0.194	0.013
Tung Chung	22.1	6.8	3.4	0.65	0.235	0.016
Central/western	21.9	6.8	3.8	0.78	0.232	0.015
Shenzhen	25.4	6.6	5.8	1.51	0.321	0.018
Zhongshan	21.7	7.1	4.4	1.29	0.249	0.018
Guangzhou	47.0	11.4	14.7	3.15	0.937	0.228
Conghua	39.9	12.4	13.0	1.44	1.617	0.119
Northerly flow						
Tap Mun	32.8	9.2	10.0	1.29	1.025	0.099
Tung Chung	39.5	9.2	13.5	2.72	1.102	0.117
Central/western	36.4	8.5	12.3	2.29	0.963	0.089
Shenzhen	52.4	10.2	19.4	4.00	1.337	0.164
Zhongshan	60.6	13.3	23.8	3.33	2.443	0.308
Guangzhou	44.2	9.9	16.4	3.89	1.250	0.163
Conghua	26.5	7.8	9.3	1.03	0.848	0.101
Mixed flow						
Tap Mun	38.9	12.3	8.5	0.94	0.592	0.071
Tung Chung	38.5	11.6	10.3	2.11	0.529	0.051
Central/western	43.4	12.0	11.1	1.93	0.537	0.061
Shenzhen	53.2	10.9	16.7	4.13	0.747	0.083
Zhongshan	62.1	15.7	19.7	3.18	0.966	0.132
Guangzhou	107.1	21.4	35.7	5.89	2.763	0.392
Conghua	46.3	11.8	16.5	1.59	1.979	0.257

transporting air masses northward from the ocean, a large difference is seen between $PM_{2.5}$ average concentrations at the southernmost five sites (Tap Mun, Tung Chung, and central/western in Hong Kong; Shenzhen and Zhongshan in Guangdong) and the northern two (Guangzhou and Conghua in Guangdong). As shown in Table 4, mean values from the southern five sites range from 18 to 25 μg m^{-3}, as compared to 47 μg m^{-3} measured at the more northerly urban Guangzhou site. The mean $PM_{2.5}$ concentration at the northern background site in Conghua is 40 μg m^{-3}, nearly twice as high as the sites south of Guangzhou. It is apparent that a source area must be located near Guangzhou to cause the observed accumulation of fine particulate mass over a relatively short distance. Comparing rural Conghua located North of Guangzhou with the Zhongshan site placed South of Guangzhou, an 18 μg m^{-3} increase representing a near doubling of downwind $PM_{2.5}$ is observed and can be attributed to sources located in the region between the two sites, including the city of Guangzhou. In addition to the contribution to downwind $PM_{2.5}$ by the Guangzhou area, it should be noted that the levels at the upwind background site of Tap Mun (18 μg m^{-3}) indicate significant region-wide background $PM_{2.5}$. To investigate further the origin of the particulate matter, 24 h back-trajectory modeling was performed on the 4 days of southerly flow, using the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph, 2003) with NCEP/GDAS FNL reanalysis meteorological data. Modeled trajectories were calculated for the two background sites, Tap Mun and Conghua, at elevations of 100, 500 and 1500 m. Although the rugged terrain of the region imparts uncertainty to meteorological modeling, the HYSPLIT model did confirm that the 4 southerly-flow days had air parcels transported inland from the ocean area to the South. This implies that southerly flow fine particulate levels at rural Tap Mun may be due to long-range transport. Local shipping emissions may also affect overall levels at Tap Mun, but the relative influence of this source requires further investigation.

Sulfate is a dominant component of fine particulate matter in the PRD, on average contributing 21–32% of overall mass, as shown in Table 2. The relative concentrations of sulfate during southerly wind are highest at the Guangdong background site at Conghua (1.09), shown in Fig. 3. As with overall $PM_{2.5}$, relative values of sulfate at the southern five sites are about half that measured in Conghua and are within a close range of one another (0.44–0.53), with absolute concentrations of 6.0–7.1 μg m^{-3}. It is apparent that a source of sulfate lies in the Guangzhou vicinity, leading to a doubling of concentrations from sites South of Guangzhou to northernmost Conghua. However, though levels of sulfate at the southern sites are far less than northern areas, the sulfate concentrations at the southernmost five sites are still substantial. With sulfate levels at remote Tap Mun similar to that at urban Shenzhen, it is expected that sulfate has regional background levels during southerly flow that results in approximately half of the PRD's sulfate mass. This background sulfate may be due to long-distance transport from outside of the PRD region.

Both primary and secondary in origin, relative concentrations of organic compounds at Conghua are more than double the values measured at the southern five sites during southerly wind patterns,

as shown in Fig. 3. The change between the southern five sites and Guangzhou is even more extreme, with a near tripling of relative concentrations. Comparing the southerly flow distributions of OM and sulfate, a higher range of OM concentrations is observed among the southernmost five sites (3.3–5.8 $\mu g\,m^{-3}$), with mean normalized concentrations also showing a wider range (0.20–0.35). Some localized influence on OM is thus predicted in the Hong Kong vicinity, with a tripling of OM concentrations in Guangzhou attributed to both transport of precursors from the South and locally emitted organic species in the Guangzhou area. The relatively high OM concentrations observed in Guangzhou are perhaps not surprising given the size of the city and intense traffic congestion.

Elemental carbon during southerly flow has a unique pattern compared with $PM_{2.5}$, sulfate and OM. Among the southern five sites, background Tap Mun has a low relative concentration of 0.13 while more developed sites at Shenzhen and Zhongshan are nearly three-fold higher with relative concentrations of 0.55 and 0.47, respectively. With normalized concentrations at the Hong Kong sites of Tung Chung and central/western more than doubling background Tap Mun and even higher increases at urban sites within Guangdong, local sources appear to strongly affect EC levels throughout the delta. Though EC concentrations seem to be mainly dominated by local sources, some impact of transport is apparent with high relative levels observed at Conghua (0.56), located downwind of Guangzhou. As with OM, the relatively high EC concentration in urban Guangzhou strongly suggests the importance of local sources on particulate matter concentrations.

As shown in Fig. 3, the southerly flow case for potassium shows extraordinarily high relative concentrations at rural site Conghua, more than doubling urban Guangzhou and higher than sites South of Guangzhou by a factor of five. A tracer for biomass burning, the significant increases moving northward throughout the delta lead to the conclusion that biomass burning sources are distributed within the northern section of the monitoring area, both near Guangzhou and North of the city toward Conghua. Comparatively low measured potassium at sites south of Guangzhou points to a lack of biomass burning in close proximity to Hong Kong and southern Guangdong.

Of all observed species, lead appears to be most significantly dominated by sources in Guangdong, with homogeneously low relative concentrations South of Guangzhou (ranging from .09 to .11) and levels at Guangzhou and Conghua higher by more than six-fold. Shown in Fig. 3, the sudden jump in lead levels moving from sites Zhongshan and Shenzhen to nearby Guangzhou indicates a localized source area of lead within the vicinity of Guangzhou and perhaps North of the city. Assuming no local production of lead near Conghua, the high lead concentrations at Conghua appear to be caused by transport from upwind Guangzhou.

4.2.2. Northerly flow

As seen in Fig. 3, northerly flow relative levels of $PM_{2.5}$ at sites South of Guangzhou more than double that observed during southerly flow, while normalized concentration at northernmost Conghua decreases by 0.2. A spatial gradient is seen among sites downwind of Guangzhou, with highest relative concentrations at Zhongshan (1.4) and Shenzhen (1.2) and lower levels in the Hong Kong area (0.75–0.93). Comparing upwind Conghua and downwind Zhongshan, an increase of 34 $\mu g\,m^{-3}$ can be linked to the Guangzhou region located in-between the two sites. With an attenuation of impact related to distance from Guangzhou, the increase in concentration at the background site of Tap Mun relative to Conghua is 6.3 $\mu g\,m^{-3}$. Overall, the doubling increase in relative concentrations at the three sites in Hong Kong as compared to southerly flow conditions points to the significant impact of the Guangzhou area on levels of fine particulate matter in Hong Kong. In addition to increases observed downwind of Guangzhou, northerly flow $PM_{2.5}$ measured upwind at rural Conghua is significantly high ($\sim$27 $\mu g\,m^{-3}$), indicating a regional background concentration that may be due to long-range transport from northern areas.

Similar to the reversal observed in $PM_{2.5}$ concentrations when comparing cases of northerly and southerly flows, particulate sulfate levels likewise increase at sites downwind of Guangzhou and decrease at Conghua, located upwind of Guangzhou. As observed in Fig. 3, Zhongshan receives the heaviest dose of sulfate, with concentrations relative to Guangzhou at 1.34 compared with 0.53 under southerly flow. Conghua now has lowest relative concentrations in the region (0.78) compared with ranking highest when downwind of Guangzhou. Assuming negligible local impact on particulate sulfate concentrations at Conghua, its northerly flow average sulfate concentration of 7.8 $\mu g\,m^{-3}$

indicates significant background particulate sulfate advected into the PRD that constitutes over half of the 13.3 $\mu g\,m^{-3}$ of sulfate measured at Zhongshan. Examining Table 4, it is interesting to note that the average difference between the maximum at Conghua and upwind Zhongshan under southerly flow is 5.3 $\mu g\,m^{-3}$, while the northerly flow difference between the same two sites is 5.5 $\mu g\,m^{-3}$. Thus, the direct contribution of the Guangzhou vicinity to particulate sulfate can be roughly estimated at 5–6 $\mu g\,m^{-3}$.

Having a nearly identical distribution as the northerly flow case of $PM_{2.5}$, OM concentrations appear to be influenced by a source area near Guangzhou. Impact based on proximity to Guangzhou is again observed, with Zhongshan and Shenzhen having much higher average OM levels relative to that seen during southerly flow and lesser increases in OM concentrations at sites in Hong Kong. Though OM concentrations appear to have a regional increase at sites in Hong Kong, a localized influence is still evident with normalized OM levels observed at rural Tap Mun ~20% lower than nearby Tung Chung and central/western, as shown in Fig. 3. With expected biomass burning sources near Conghua, as indicated by high potassium levels during southerly flow, OM concentrations at Conghua may be likewise influenced by nearby sources and thus not indicative of regional background levels. Even with the possible presence of OM sources near Conghua, the impact of transported organic particulate species from Guangzhou is significant. Absolute OM concentrations displayed in Table 4 show a northerly flow difference between downwind Zhongshan and upwind Conghua of 14.5 and an 8.5 $\mu g\,m^{-3}$ increase from Zhongshan to Conghua during southerly flow.

With much lower relative concentrations at background sites (Tap Mun, Conghua) compared with urban areas (Shenzhen, Guangzhou), the distribution of EC during northerly flow appears to be dominated by local sources. However, it should be pointed out that some degree of transport is seen in concentrations at the background sites. Comparing the case of northerly winds to that of southerly winds, relative EC concentrations at Tap Mun increase by 0.21 during flow from the North, while normalized levels at Conghua decrease by 0.30, as shown in Fig. 3. Despite the observed transport of EC, local influence appears to remain significant at the three sites in Hong Kong, with much higher EC concentrations at central/western and Tung Chung (2.3 and 2.7 $\mu g\,m^{-3}$, respectively) as compared with background Tap Mun (1.3 $\mu g\,m^{-3}$).

While normalized potassium at Conghua is seen to double levels at Guangzhou during southerly flow, as shown in Fig. 3, Conghua ranks lowest among all sites during wind from the North, indicating significant biomass burning occurring South of Conghua. The reversal of flow causes highest relative levels at Zhongshan (1.9) with lessening impact moving southward to the sites in Hong Kong (.78–.91). Though Conghua has the lowest potassium concentration during the case of northerly flow, the level is still relatively high (0.85 $\mu g\,m^{-3}$) and indicates a background contribution that constitutes more than a third of the peak level observed at Zhongshan. The background potassium during northerly flow may be caused by biomass burning located near Conghua or due to transport from North of the PRD region.

Normalized particulate lead concentrations are observed to dramatically increase at sites south of Guangzhou when comparing the case of northerly flow to southerly flow, with a maximum 20-fold increase observed at Zhongshan and a minimum five-fold increase observed at central/western. With relative concentrations spiking at Zhongshan (2.0), attenuation is again seen moving southward to Shenzhen (1.0) and the sites in Hong Kong (0.56–0.71). While absolute lead concentrations, shown in Table 3, are observed to increase from 0.01 to 0.10 $\mu g\,m^{-3}$ at the background site of Tap Mun, comparing cases of flow from the North and South, only a slight decrease of 0.02 $\mu g\,m^{-3}$ in concentration is seen at Conghua. It is expected that particulate lead is regionally advected into the PRD during flow from the North, maintaining high concentrations of lead at Conghua. However, local sources of lead at the background site in Guangdong cannot be ruled out. Even given a rise in background levels of lead during northerly flow, a tripling in absolute lead concentrations from Conghua to Zhongshan indicates lead emissions local to Guangzhou.

4.2.3. *Mixed flow*

Even though larger rainfall was observed during days within the mixed flow category in comparison with northerly and southerly flows, the stagnant conditions result in extremely high $PM_{2.5}$ concentrations at Guangzhou, with levels more than 60 $\mu g\,m^{-3}$ higher than that observed for the other two flow categories, as presented in Table 4. In

comparison to the dramatic change observed in Guangzhou, a more muted increase is seen at the remainder of sites in the delta, resulting in relative fine particulate levels at Guangzhou more than 40% higher than any other sampling site. This observed maximum at Guangzhou is similarly observed for all presented species (sulfate, OM, EC, potassium and lead). With limited transport of fine particulate concentrations, it is observed that sources within the vicinity of Guangzhou heavily impact local pollution during mixed flow. Throughout the delta, stagnant winds result in an accumulation of fine particulate matter from both local and regional sources, leading to higher $PM_{2.5}$ concentrations at both background sites at Conghua ($46\,\mu g\,m^{-3}$) and Tap Mun ($39\,\mu g\,m^{-3}$) than observed during southerly or northerly flows. The relatively low wind speeds likely favor more local influences at the sampling stations.

Compared with all other examined species, the mixed flow distribution of normalized sulfate concentrations indicates the most significant degree of regional impact, with high relative concentrations (0.53–0.74) observed at the six sites surrounding Guangzhou. In contrast, the same six sites have lower relative levels of $PM_{2.5}$ (0.36–0.58) as shown in Fig. 3. Though sulfate sources appear to have a regional influence on concentrations in the PRD, sources in the vicinity of Guangzhou city cause sulfate concentrations at Guangzhou to measure more than 25% higher than any other site in the region.

With a nearly identical distribution as relative concentrations of $PM_{2.5}$, normalized OM has a maximum at Guangzhou during stagnant conditions, significantly higher than the remainder of sites (ranging from 0.25 to 0.55). In comparison to the distribution of sulfate among the seven monitoring sites during mixed flow, OM levels appear to have a more localized impact. Major sources of OM are expected to be located near Guangzhou, causing a doubling of average OM concentrations at Guangzhou in comparison with southerly and northerly flows, despite higher precipitation during mixed flow days. Although a doubling in absolute OM is observed at Guangzhou, comparing mixed flow to northerly flow, a 10–20% decrease is observed at the southernmost five sites, indicating an isolation of high OM levels to Guangzhou.

As observed during northerly and southerly flows, a localized influence on EC is again apparent during mixed flow, with relative concentrations at the urban areas of Guangzhou and Shenzhen (0.69) much higher than background levels at Conghua (0.29) and Tap Mun (0.18). Localized sources are also apparent within the Hong Kong region, with normalized EC at central/western and Tung Chung more than doubling that measured at background Tap Mun.

The distribution of potassium concentrations during mixed flow supports a source area located near Guangzhou and Conghua. Ranked highest in the region during stagnant conditions, levels of potassium at Guangzhou ($2.8\,\mu g\,m^{-3}$) are more than double concentrations measured during both southerly and northerly flows, in spite of higher rainfall during mixed flow days. Though potassium levels increase at Conghua by 130%, comparing the case of mixed flow to northerly flow, a decrease of 40–60% is measured among the five sites South of Guangzhou. The stagnant winds appear to isolate high concentrations of potassium to the northern PRD region and indicate a source area affecting concentrations at both Guangzhou and Conghua.

With a nearly identical distribution as particulate potassium, levels of lead during mixed flow similarly appear to be dominated by sources in the northern PRD and have little transport to sites south of Guangzhou. Compared with northerly flow, the southernmost five sites have a 30–60% reduction in absolute levels of lead while Guangzhou and Conghua increase by 140% and 160%, respectively. With mixed flow lead concentrations at remote Tap Mun ($0.07\,\mu g\,m^{-3}$) similar to that at urban Shenzhen ($0.08\,\mu g\,m^{-3}$), particulate lead in Hong Kong vicinity appears to be regionally controlled.

5. Conclusions

Development of an effective fine particulate management plan in the Pearl River Delta has been hindered by a lack of information about the regional nature of $PM_{2.5}$, with regional chemical composition, influencing source areas and meteorological impacts yet unknown. To assess fine particulate pollution throughout the PRD, simultaneous 24 h filter measurements were conducted at seven sites during October 2002, December 2002, March 2003 and June 2003. Combining the 4 months of sampling, overall average $PM_{2.5}$ concentrations at all sites far exceed the United States national ambient air quality standard of $15\,\mu g\,m^{-3}$ (annual average), with levels in Hong Kong ranging from 29 to $34\,\mu g\,m^{-3}$ and even higher average

concentrations in Guangdong ranging from 37–71 $\mu g\,m^{-3}$. Despite the variability in concentrations throughout the region, the general chemical make-up of the fine particulate matter is very similar among the seven sites, with organic mass and sulfate dominating fine particulate mass at 24–35% and 21–32%, respectively. The significant levels of organic mass and sulfate, both related to fossil fuel combustion, points to the impact of anthropogenic activities on local air quality. Other measured species include crustal matter (7–13%), ammonium (6–8%), elemental carbon (3–8%) and nitrate (1–6%). To our knowledge, these are the first reported measurements of overall chemical composition of fine particulate matter in Guangdong.

Combined analysis of local meteorology (wind speed, wind direction and precipitation) and fine particulate levels illustrates the significant influence wind patterns have on regional air quality. The impact of transport was investigated by categorizing 13 (of 20 total) sampling days by prevailing wind direction (southerly, northerly and low-speed mixed flows). Comparison of $PM_{2.5}$ levels at sites immediately upwind and downwind of Guangzhou during northerly and southerly flow conditions indicates an estimated contribution of 18–34 $\mu g\,m^{-3}$ to downwind $PM_{2.5}$ by sources in the vicinity of Guangzhou city. A gradient effect was observed, with the most extreme increases in fine particulate matter during northerly winds occurring at Zhongshan, located close to Guangzhou and lesser change observed at the more distant sites.

Looking into the impacts of wind patterns on the spatial distribution of specific fine particulate species, contributing source regions can be assessed in greater detail. Sulfate, related to the burning of coal, has a high regional background level estimated at 6–8 $\mu g\,m^{-3}$, over half of the total measured sulfate. Analysis of sulfate concentrations at sites upwind and downwind of Guangzhou indicates a direct input of 5–6 $\mu g\,m^{-3}$ from sources near Guangzhou city. Guangzhou also stands out as a prominent regional source of organic mass (OM), with observed increases of 8.5–14.5 $\mu g\,m^{-3}$ at sites immediately downwind and a disproportionate elevation in organic mass at the Guangzhou site during stagnant conditions. Local OM sources are also evident in the Hong Kong region, with rural Tap Mun consistently lower than urban central/western. regional levels of elemental carbon (EC) are highest during all flow conditions at urban Shenzhen and Guangzhou. Local sources of EC are evident within Hong Kong, with more developed sites at Tung Chung and central/western having nearly double the EC concentrations measured at rural Tap Mun. In contrast, the distribution of potassium (biomass burning) and lead (industrial sources, combustion of fossil fuels) indicate significant sources in northern area of the delta influencing concentrations downwind. The regional distribution of potassium points to sources in the vicinity of both Guangzhou and Conghua, with strikingly high levels observed at Conghua during southerly flow. Regional levels of lead appear to be controlled by sources in the vicinity of Guangzhou.

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