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Long-term trends of ambient particulate matter emission source contributions and the accountability of control strategies in Hong Kong over 1998–2008

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HIGHLIGHTS

- Positive Matrix Factorization is applied to identify PM_{10} sources in Hong Kong.
- Local and non-local PM_{10} sources are classified based on spatiotemporal variations.
- Local control measures on vehicle exhaust are effective with more than 50% reduction.
- Local reduction is totally offset by increased contributions from non-local sources.
- A coordinated, regional-scale air quality management plan is urgently needed.

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ABSTRACT

Despite extensive emission control measures targeting motor vehicles and to a lesser extent other sources, annual-average PM_{10} mass concentrations in Hong Kong have remained relatively constant for the past several years and for some air quality metrics, such as the frequency of poor visibility days, conditions have degraded. The underlying drivers for these long-term trends were examined by performing source apportionment on eleven years (1998–2008) of data for seven monitoring sites in the Hong Kong PM_{10} chemical speciation network. Nine factors were resolved using Positive Matrix Factorization. These factors were assigned to emission source categories that were classified as local (operationally defined as within the Hong Kong Special Administrative Region) or non-local based on temporal and spatial patterns in the source contribution estimates. This data-driven analysis provides strong evidence that local controls on motor vehicle emissions have been effective in reducing motor vehicle-related ambient PM_{10} burdens with annual-average contributions at neighborhood- and larger-scale monitoring stations decreasing by $\sim$6 $\mu g\ m^{-3}$ over the eleven year period. However, this improvement has been offset by an increase in annual-average contributions from non-local contributions, especially secondary sulfate and nitrate, of $\sim$8 $\mu g\ m^{-3}$ over the same time period. As a result, non-local source contributions to urban-scale PM_{10} have increased from 58% in 1998 to 70% in 2008. Most of the motor vehicle-related decrease and non-local source driven increase occurred over the period 1998–2004 with more modest changes thereafter. Non-local contributions increased most dramatically for secondary sulfate and secondary nitrate factors and thus combustion-related control strategies, including but not limited to power plants, are needed for sources located in the Pearl River Delta and more distant regions to improve air quality conditions in Hong Kong. PMF-resolved source contribution estimates were also used to examine differential contributions of emission source categories during high PM episodes compared to study-average behavior. While contributions from all source categories increased to some extent on high PM days, the increases were disproportionately high for the non-local sources. Thus, controls on emission sources located outside the Hong Kong Special Administrative Region will be needed to effectively decrease the frequency and severity of high PM episodes.

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1. Introduction

During the past twenty years, the environmental authorities in Hong Kong have undertaken a series of air pollution control measures including but not limited to requirements for low-sulfur diesel fuel, lead-free gasoline, three-way catalytic converters, conversion of diesel taxis and minibuses to liquefied petroleum gas, and capping the VOC content of some solvents and consumer products. Despite these measures, the long-term trends for various ambient air quality metrics show either no improvement or a worsening of conditions. For example, the open circles in Fig. 1 show the annual-average PM_{10} mass concentration composited over six monitoring stations that collectively represent urban-scale conditions (these sites are described later in this paper). Hourly data from the PM_{10} monitoring network were used to construct the annual averages. Annual-average PM_{10} mass was relatively constant over the period from 1998 to 2008 with a 9% increase in mass for the five years starting in 2004 compared to the five year period starting in 1998. The solid circles in Fig. 1 show the percentage of hours with poor visibility (visibility less than 8 km with relative humidity less than or equal to 80%) measured at the Hong Kong Observatory (HKO, 2012). The frequency of poor visibility days increased throughout the 1990s with more dramatic increases during first half of the 2000s. A local maximum was observed in 2004 (18% of hours) with the frequency of poor visibility hours decreasing through 2008 and then increasing again through 2011. The frequency of hours with poor visibility increased by 47% for the five years starting in 2004 compared to the five year period starting in 1998. The substantially greater worsening of visibility compared to the more modest increase in PM_{10} mass has been driven by changes in chemical composition of the PM_{10} aerosol. Fig. 1 also shows that over the period from 1998 to about 2004, for the same six sites used to construct the PM_{10} TEOM mass trend, the annual average total carbon dramatically decreased while annual average sulfate significantly increased. Thus, while there was little change in PM_{10} mass during this period, the composition shifted towards an aerosol with higher mass extinction efficiency that leads to a reduction in visibility (Andreae et al., 2008).

The two most pervasive air pollution issues in Hong Kong are street-level (especially near roadway) air quality and regional-scale air quality that affects not only Hong Kong but also the entire Pearl River Delta (PRD). The emission inventory reveals gasoline and diesel vehicles to be the main sources of street-level pollution (Zheng et al., 2009a). Regional-scale air pollution, however, is caused by the cumulative impacts from numerous emission source categories such as motor vehicles, marine vessel emissions, industry and power plants located in Hong Kong, the greater PRD region, and beyond. As shown in Fig. 2, the PRD region of Guangdong Province, which historically was Hong Kong's hinterland, has become highly urbanized and indeed has developed as the largest workshop in the world. Over the past fifteen years, pollutant emissions within the PRD region have increased dramatically as a result of booming industrial activities and relatively modest pollution control regulations. Therefore, the pollutants transported from the greater PRD region to Hong Kong are expected to have correspondingly increased.

Effective air quality management necessarily requires setting priorities and therefore it is important to identify the emission source categories leading to deterioration of air quality in Hong Kong and quantify the long-term trends in the contributions from such sources. To the extent there are large contributions from within the Hong Kong Special Administrative Region (HKSAR) the local government can implement additional controls on. However, if the dominant contributions are from the greater PRD area and more distant regions, a regional planning process that involves cooperation between Hong Kong, Guangdong Province and perhaps other provinces in Mainland China will be necessary to efficiently manage air quality in Hong Kong.

This study was commissioned to extend our previous work to apportion ambient PM_{10} levels in Hong Kong (Yuan et al., 2006; Huang et al., 2009) with emphasis on further identifying and quantifying the drivers for the long terms trends in Hong Kong air quality such as those features demonstrated in Fig. 1. Insights were sought into whether air quality benefits from prior control measures could be elucidated from routine monitoring data. This information could be used to frame and prioritize future control policies. Following the methodology of the previous studies, receptor modeling was performed on the PM_{10} speciation data.

2. Data collection and analysis

For more than twenty years, the Hong Kong Environmental Protection Department (HKEPD) has operated a PM_{10} chemical speciation network (network) that has included several

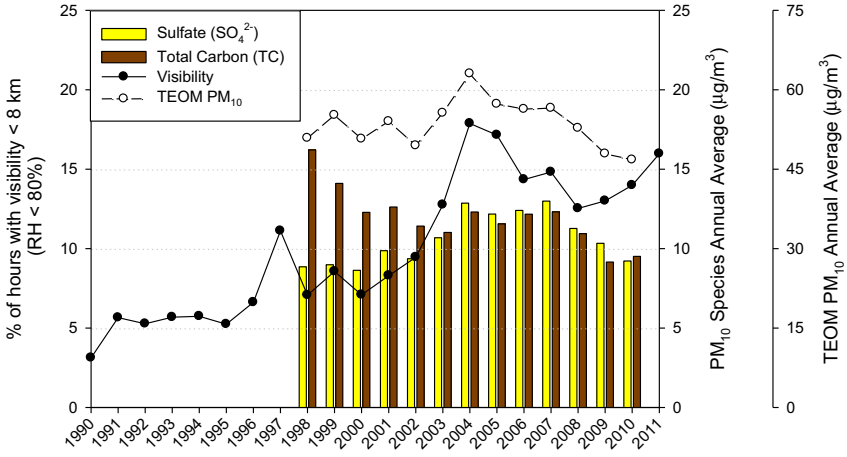

Fig. 1. Historical trends for visibility impairment and PM_{10} mass, sulfate and total carbon. Data sources and calculation methodology are described in the text.

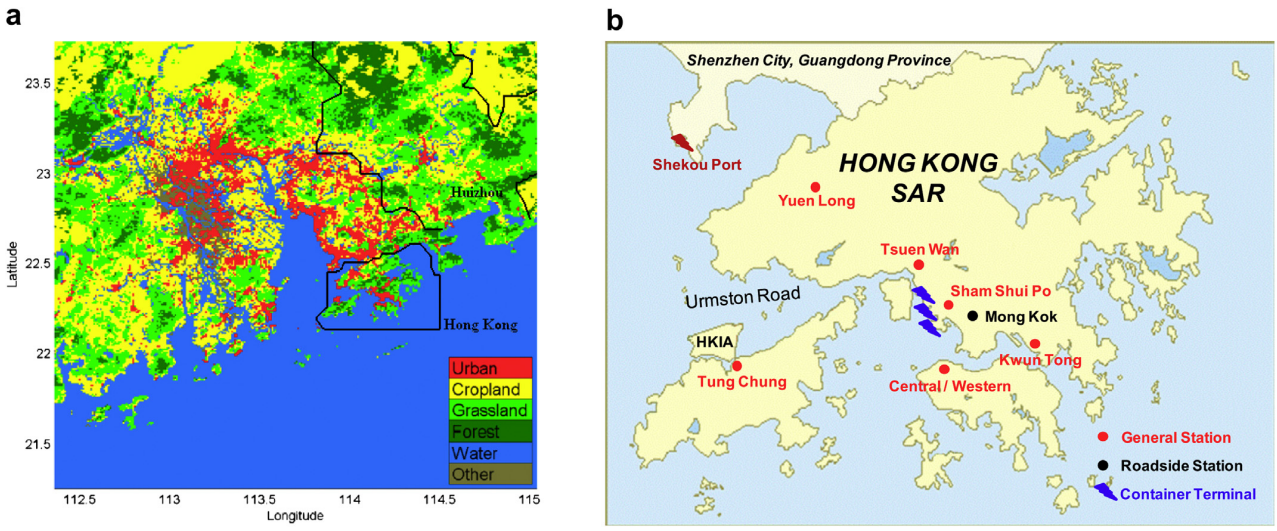

Fig. 2. (a) Land-use map of the Pearl River Delta in 2003, and (b) geographical distribution of the Air Quality Monitoring Stations in the Hong Kong Special Administrate Region.

monitoring stations throughout the HKSAR. 24-h integrated (midnight-to-midnight) samples are collected on a 1-in-6 day frequency using High Volume PM_{10} samplers with quartz filters. The filters are analyzed for gravimetric mass, elements by inductively coupled plasma atomic emission spectroscopy (ICP-AES) and ions by ion chromatography (IC). Since January 1998, a thermal/optical transmittance method following the NIOSH 5040 protocol (Chow et al., 2001) has been used to obtain organic carbon (OC) and elemental carbon (EC). Table 1 summarizes the valid sampling periods and environmental characteristics for each of the seven stations that have operated nearly continuously over the 1998–2008 period and were used in this study. The first six stations (YL through KT) are neighborhood-scale sites and collectively are considered to represent urban-scale PM_{10} levels albeit with some underlying spatial variability. For the purpose of this study they are termed "general stations" and this descriptor is used when analyses are presented as the average over these six sites. General stations are typically located on rooftops of four- to six-story buildings with unobstructed airflow from most directions. The remaining site is the MK station that is located only a few meters above the ground in a heavily trafficked area to gauge roadside exposures by pedestrians. Towards the end of 2000, the MK station was relocated to its current location and started operation on 2 January 2001. This relocation caused a discontinuity in the time series for PM_{10} mass and some species. Therefore, data for MK is included only for the period from 2001 onward. Detailed information about the network can be found in Yuan et al. (2006) and Section 1 of Supplementary information.

Source apportionment modeling was performed using Positive Matrix Factorization (PMF). PMF is a factor analytic method that provides a parameterized convergence scheme and constrains all the elements in the factor score and factor loading matrices to be positive (within a narrow tolerance). These features are attractive for the modeling of environmental data sets and PMF has been widely used for the source apportionment of different ambient pollutants, including particulate matter (e.g. Lee et al., 1999; Lee et al., 2003), volatile organic compounds (e.g. Song et al., 2007; Lau et al., 2010), polycyclic aromatic hydrocarbons (e.g. Larsen and Baker, 2003), and organic aerosols (e.g. Lanz et al., 2007), and also of aerosol size distribution (e.g. Zhou et al., 2005). In this study, PMF modeling was performed using EPA PMF Version 3.0 (USEPA, 2008).

The goal of PMF applied to ambient particulate matter data sets is to resolve for a specified number of factors (p), the factor loadings (f_{kj} for species j and factor k, which operationally are species mass fractions in the source profiles) and factor scores (g_{ik} for sample i and factor k, which operationally are sample-specific source contribution estimates) -

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \quad (1)$$

where x_{ij} is the element in the species ambient concentration matrix with n the number of samples, m the chemical species measured, and e_{ij} is the residual for the element corresponding to the ith sample and the jth species. The PMF solution minimizes the objective function Q using data point-specific uncertainties u_{ij} -

Table 1
Monitoring station characteristics.

Station		Count	Start date	End date	Major data gap	Station characteristics
Yuen Long	YL	654	Jan. 2, 1998	Dec. 26, 2008		Urban/Residential
Tung Chung	TC	575	Apr. 3, 1999	Dec. 28, 2008		New Town/Residential
Tsuen Wan	TW	631	Jan. 3, 1998	Dec. 26, 2008	Feb. 6, 2003–Apr. 23, 2003	Urban/Residential/Commercial
Sham Shui Po	SSP	586	Jan. 11, 1998	Dec. 26, 2008	Nov. 29, 2003–Mar. 16, 2004; May 12, 2007–Jun. 5, 2008	Urban/Commercial
Central/Western	CW	641	Jan. 2, 1998	Dec. 26, 2008	Nov. 9, 2001–Mar. 27, 2002	Urban/Residential
Kwun Tong	KT	598	Jan. 4, 1998	Dec. 31, 2008	Oct. 31, 1998–Jan. 11, 1999; Apr. 13, 2002–Feb. 1, 2003	Commercial/Residential/Near ferry pier
Mong Kok	MK	482	Jan. 2, 2001	Dec. 27, 2008		Roadside, relocated at the end of 2000

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\left[\frac{x_{ij} - \sum_{k=1}^{p} g_{ik}f_{kj}}{u_{ij}}\right] \quad (2)$$

It is therefore important to provide plausible uncertainty estimates that can reflect the variability and artifact in the processes of sampling, chemical analysis and transformation of pollutant profiles in the air.

In this study, concentration dependent error structures were assumed with the form $U_{ij} = a_j + b_j \times C_{ij}$ where U_{ij} is the uncertainty for species j in sample i, C_{ij} is the mass concentration of species j in sample i, and a_j and b_j are the species-specific additive and multiplicative contributions, respectively, to the uncertainty. Uncertainty estimates were conditioned using the methodology of Polissar et al. (1998). For each species an effective minimum detection limit (MDL, denoted as d_j), was estimated as three times a_j which is the additive term of the error structure. For concentration values below the MDL the uncertainty was set to $U_{ij} = 5d_j/6 = 5a_j/2$. For each species the a_j and b_j coefficients were derived from an analysis of collocated samples routinely collected at the SSP station and evaluated using collocated samples also routinely collected at the TW station. The methodology to develop the error structures from collocated data is summarized in Section 2 of the Supplementary information. Table 2 lists the derived a_j and b_j values, effective MDL and percentage of samples below the effective MDL for each species.

Data from all sites were combined into a single dataset of 4167 samples to generate a consistent set of source profiles across the sites. The quartz fiber filter gravimetric mass (hereafter denoted as QTM) was used as the total mass variable. The rationale for using QTM as the total mass variable is discussed in Section 3 of the Supplementary information. 10% extra modeling uncertainty was imposed to account for possible temporal changes in the source profiles and other sources of variability. Twenty base runs were performed and the run with the minimum Q value was selected as the base run solution. 100 bootstrap runs were conducted with minimum correlation R-value of 0.6 to examine the stability and to estimate the uncertainty of the base run solution. All of the bootstrapped factors were uniquely mapped to a factor from the base solution, and no factors were unmapped. For the Q values derived from the twenty base runs, the standard deviation was only ~0.004% of the mean, indicating a very stable solution. *FPEAK* is a parameter within EPA PMF to control the rotations. A range of *FPEAK* values were examined and an *FPEAK* value of zero, indicating no additional control over the rotation, was selected for this modeling. Modeling was performed for four to ten factors and the nine factor solution was deemed to be most representative, as discussed in detail in Section 4 of the Supplementary information.

Putative emission source categories were mapped onto the factors by identifying the tracer(s) for each source which are typically the species that exclusively or largely reside in a particular source. Table 3 lists the emission source categories and corresponding tracers used for matching PMF-resolved factors to the sources. It is noted that the bootstrap method accounts for some but not all sources of uncertainty and thus is likely a lower bound on the true uncertainties in the modeling results. The PM_{10} emission source categories assigned to the nine factors are vehicular exhaust, residual oil combustion, fresh sea salt, aged sea salt, crustal material/soil, secondary sulfate, secondary nitrate, trace metals from sources such as zinc smelting, and coal combustion admixed with biomass burning, Detailed information on source identification can be found in Yuan et al. (2006) and Section 4 of the Supplementary information.

To examine the robustness of the PMF source apportionment results, additional receptor models including principal components analysis with absolute principal components scores (APCA, Thurston and Spengler, 1985) and Unmix (USEPA, 2007) were also applied. Generally, the three models yielded similar estimations for sources of vehicle exhaust, residual oil, crustal soil, secondary sulfate, and coal combustion/biomass burning. Considering that APCA and Unmix cannot resolve distinct aged sea salt and secondary nitrate factors and significant negative loadings onto a few factors were observed, the PMF results were considered the most reasonable and used for the remainder of the study. APCA and Unmix results are provided in Section 5 of the Supplementary information for reference.

Table 2
Additive (a_j) and multiplicative (b_j) error terms derived using collocated data, effective minimum detection limit (defined in Section 2), and the percentage of samples below the effective detection limit.

Species j	a_j (μg m^{-3})	b_j	Effective MDL (μg m^{-3})	% of samples below effective MDL
Al	0.046	0.071	0.014	1%
As	0.0001	0.050	0.0002	25%
Ca	0.022	0.043	0.065	0%
Cd	0.00002	0.058	0.0001	28%
Cl^-	0.014	0.047	0.042	0%
Fe	0.004	0.030	0.013	0%
K^+	0.012	0.028	0.035	0%
Mg	0.005	0.020	0.015	0%
Mn	0.0002	0.029	0.001	27%
Na^+	0.026	0.018	0.079	0%
NH_4^+	0.017	0.028	0.051	0%
Ni	0.0003	0.050	0.0008	36%
NO_3^-	0.043	0.026	0.129	0%
Pb	0.0002	0.035	0.001	20%
SO_4^{2-}	0.060	0.025	0.179	0%
V	0.00001	0.047	0.00002	48%
Zn	0.002	0.032	0.005	6%
EC	0.115	0.023	0.344	0%
OC	0.112	0.032	0.335	0%

3. Results and discussion

3.1. Source contributions and their seasonal and spatial variations

As previously discussed, one objective of the source apportionment modeling was to determine local (again, operationally identified as sources within the HKSAR) and non-local emission source categories and their respective contributions because the control of local sources is fully within Hong Kong's jurisdiction while controlling non-local sources requires regional cooperation. The approach largely focused on assigning sources as local or non-local based on seasonal and spatial patterns in the PMF-modeled source contribution estimates (SCE).

Table 3
Tracers applied for source identification and assignment of factors as primarily of local (within the HKSAR) or non-local origin.

Source	Tracers	Dominant spatial scale
Vehicle exhaust	EC, OC	Local
Residual oil	Ni, V	Local
Fresh sea salt	Na^+, Mg^{2+}, Cl^-	Local
Aged sea salt	Na^+, Mg^{2+}, NO_3^-, SO_4^{2-}	Local
Crustal soil/Dust	Al, Ca, Mg, Fe	Non-local
Secondary sulfate	NH_4^+, SO_4^{2-}	Non-local
Secondary nitrate	NH_4^+, NO_3^-	Non-local
Trace metals	Zn	Non-local
Coal combustion/Biomass burning	As, Cd, Pb, K^+, OC	Non-local

Hong Kong is situated in the subtropics along the southeast coast of Eurasia Continent. The seasonal evolution of weather in Hong Kong is closely related to and controlled by seasonal evolution of the East Asian Monsoon system. In summer, a large surface low-pressure system develops over Asia in response to rising surface temperature over the continent. Consequently, cyclonic winds flow into the Asian landmass in the lower levels, leading to southerly or southwesterly prevailing winds in Hong Kong bringing clean oceanic air mass from the sea. In winter the temperatures inland become lower than that of the ocean, leading to anti-cyclonic flow over the Asian landmass. This wintertime pattern drives mainly northerly or northeasterly surface and synoptic winds that transport particulate matter to Hong Kong from the PRD and regions further inland. In light of these characteristics, seasonal and spatial patterns for the receptor modeled SCEs were examined from the perspective that local source SCEs typically should have relatively larger spatial variation over the network and smaller seasonal variation, while non-local sources should exhibit smaller spatial variation and larger seasonal variation. Based on the prevailing synoptic systems, for this analysis the seasons in Hong Kong were defined as summer from 16 May to 15 September; winter from 16 November to 15 March of following year; and a transition season, including both spring and fall, from 16 March to 15 May and from 16 September to 15 November.

Table 4 lists the annual-, wintertime-, summertime-, and transition season- averaged source contributions at general stations and roadside station during the 11-year period from 1998 to 2008. Based on the general station SCEs, the secondary sulfate factor is the largest contributor to PM_{10} mass and accounts for 22% of the period-average ambient PM_{10} in Hong Kong. SCEs for vehicle exhaust, aged sea salt, crustal soil, secondary nitrate, and coal combustion/biomass burning are comparable with each emission source category responsible for ~12–15% of the PM_{10} mass. Contributions from residual oil, fresh sea salt, crustal soil, and trace metals are generally below 5%.

Seasonal and spatial variability in the SCEs are presented in Fig. 3. The results in both Table 4 and Fig. 3 show that for sources such as vehicle exhaust, residual oil, fresh sea salt, and aged sea salt, the summertime contributions are comparable to or even slightly higher than the wintertime contributions. The weak seasonal dependence and the general lack of emission sources in the summertime upwind direction (i.e. SE/SW) leads to these sources being characterized as local. On the other hand, general stations SCEs for the crustal/soil, secondary sulfate, secondary nitrate, trace metals and coal combustion/biomass burning, emission source categories were 3–5 times higher in winter than in summer. With prevailing surface winds from NW/NE in winter bringing air masses from the PRD and regions further inland to Hong Kong, these sources are presumed to be non-local (i.e. from the PRD and larger regional scales). The assignment of emission source categories as being predominantly local or non-local is summarized in Table 3. Sources that were assigned as local based on seasonal behavior also show relatively large spatial variation across the network. In particular, the vehicle exhaust SCE at MK is nearly 2.5 times that at the general stations. Non-local sources, such as crustal soil, secondary sulfate, secondary nitrate, and trace metals, and coal combustion/biomass burning, show relatively consistent SCEs across the network. These spatial patterns are another indicator for the geographical nature of each emission source category. However, when the general stations are ordered from northwest to southeast (Fig. 3), a spatial gradient in the SCEs is discernible for most of the non-local sources with SCEs decreasing with increasing distance from the PRD. This pattern suggests that some of the non-local sources may be sufficiently close to Hong Kong to result in a spatial gradient in source contributions across the network. In contrast, such spatial gradients were not observed for the local sources.

Having classified the geographic nature of each emission source category, the relative contributions of local and non-local sources to ambient PM_{10} levels observed in Hong Kong were calculated. For the general stations, non-local sources contributed about 67% and local sources contributed about 32% to the grand-average PM_{10} during the period from 1998 to 2008. Non-local contributions exhibited relatively higher contributions in winter (77%) compared to summer (48%). At the roadside station, non-local sources contributed about 58% and local sources contributed about 44% to the PM_{10} burdens from 2001 to 2008 (there was a 2% over prediction in the estimates). Non-local contributions were 69% in winter and 36% in summer.

3.2. Long-term (eleven-year) trends in annual source contribution estimates

Inter-annual trends in SCEs over the eleven-year study period were examined for the general stations and the roadside station. Fig. 4 shows the trends in annual average SCEs for each of the nine resolved factors. Vehicle exhaust annual-average SCEs monotonically decreased over the six year period from 1998 to 2003 at the general stations with similar behavior at the roadside station for 2001 to 2003. Despite small rebounds in 2004 and 2006, the

Table 4
Annual-, wintertime-, summertime-, and transition season- averaged source contribution estimates (unit: μg m^{-3}) and contribution percentages for general stations from 1998 to 2008 and for the roadside station from 2001 to 2008 (unit: μg m^{-3}).

	General stations (YL, TC, TW, SSP, CW & KT)					Roadside station (MK)				
	Annual	Winter	Summer	Transition	Win/Sum	Annual	Winter	Summer	Transition	Win/Sum
Vehicle exhaust	8.5 (15%)	8.2 (11%)	8.9 (25%)	8.4 (15%)	0.92	21.3 (30%)	19.0 (22%)	23.3 (47%)	21.4 (28%)	0.82
Residual oil	0.2 (0.4%)	0.2 (0.3%)	0.3 (1%)	0.2 (0.4%)	0.74	0.3 (0.4%)	0.2 (0.2%)	0.3 (1%)	0.3 (0.4%)	0.57
Fresh sea salt	1.8 (3%)	2.1 (3%)	1.8 (5%)	1.4 (2%)	1.2	2.3 (3%)	2.5 (3%)	2.8 (6%)	1.7 (2%)	0.90
Aged sea salt	7.1 (13%)	6.2 (8%)	6.5 (19%)	8.6 (15%)	0.95	7.2 (10%)	6.5 (7%)	6.5 (13%)	8.5 (11%)	1.0
Crustal soil/Dust	6.7 (12%)	9.7 (13%)	3.2 (9%)	7.3 (13%)	3.0	7.5 (11%)	10.3 (12%)	4.0 (8%)	8.4 (11%)	2.6
Secondary sulfate	12.3 (22%)	16.1 (22%)	6.9 (20%)	14.1 (25%)	2.3	13.4 (19%)	16.4 (19%)	6.9 (14%)	17.1 (22%)	2.4
Secondary nitrate	8.3 (15%)	14.9 (20%)	3.1 (9%)	7.2 (13%)	4.8	11.0 (16%)	18.2 (21%)	4.5 (9%)	10.6 (14%)	4.0
Trace metals	2.6 (5%)	4.1 (5%)	1.0 (3%)	2.9 (5%)	4.0	2.8 (4%)	4.1 (5%)	0.9 (2%)	3.4 (4%)	4.4
Coal combustion/Biomass burning	7.3 (13%)	12.7 (17%)	2.6 (7%)	7.1 (12%)	5.0	6.2 (9%)	10.4 (12%)	1.7 (3%)	6.7 (9%)	6.1
Local sources	17.6 (32%)	16.7 (22%)	17.4 (50%)	18.6 (32%)	1.0	31.0 (44%)	28.2 (33%)	32.9 (66%)	32.0 (41%)	0.86
Non-local sources	37.2 (67%)	57.6 (77%)	16.8 (48%)	38.6 (67%)	3.4	41.0 (58%)	59.3 (69%)	18.1 (36%)	46.3 (60%)	3.3
Residual[a]	0.5 (1%)	0.6 (1%)	0.8 (2%)	0.2 (0.3%)		−1.1 (−2%)	−0.9 (−1%)	−1.2 (−3%)	−1.1 (−1%)	
Total	55.3	74.8	35.1	57.4		70.9	86.6	49.7	77.1	

[a] Residual refers to particle mass that is not apportioned to the PMF-resolved factors, i.e. the total mass in the residual matrix.

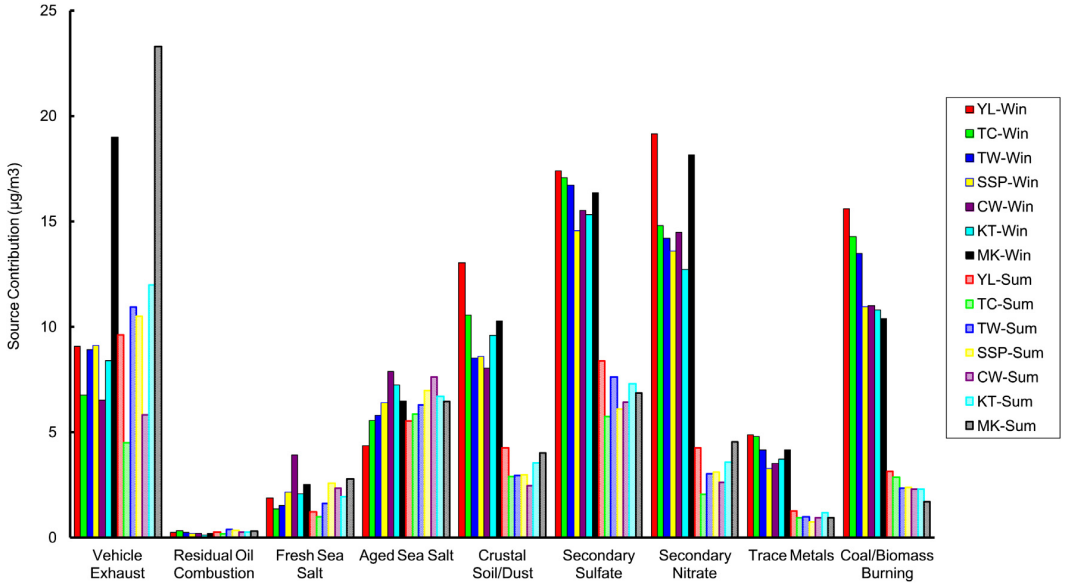

Fig. 3. Site-specific PMF-resolved summer (XX-Sum) and winter (XX-Win) source contribution estimates (μg m^{-3}) across the network, with general stations (YL to KT) from northwest to southeast (i.e., in the order of increasing distance from PRD region).

vehicle exhaust annual average SCEs at the general stations continued to decrease through 2008. This decreasing trend demonstrates the efficacy of vehicular emission control measures adopted in Hong Kong. For the general stations, the annual average contributions from vehicles were ~5 μg m^{-3} in 2008 which corresponds to a 54% reduction from the contribution of nearly 12 μg m^{-3} in 1998. Reasons for the modest SCE increases in 2004 and 2006 are not known; they might arise from year-to-year variations in atmospheric ventilation that affect pollutant dispersion or from increases in emissions in response to economic changes or other activity-based drivers. For example, over the period 1998–2007 the Hong Kong Gross Domestic Product was lowest in 2003 (HKEPD, 2008) and thus 2004 was a rebound year in economic activity. Also, the HKSAR PM emission inventory shows a monotonic decrease in annual PM emission road transport emissions from 1998 to 2007 with annual reductions averaging ~300 tonnes $year^{-1}$ (HKEPD, 2008). However, the smallest emission reduction was in 2006 (50 tonnes $year^{-1}$) which coincides with one of the years with a modest increase in the motor vehicle exhaust SCE.

Fig. 5 shows that for the years from 1998 to 2007, there is a high correlation between PMF-modeled vehicle SCEs at the general stations and the annual PM emissions from road transport estimated by HKEPD (2008). The correlation coefficient of 0.90 increases to 0.99 if data from 2004 and 2006 are excluded (the emission inventory was not available for 2008). This high correlation adds confidence to the assignment of the motor vehicle factor and its long term trend. When the PM emissions from road transport decreased by 1000 tonnes $year^{-1}$, the annual average urban-scale ambient PM_{10} burden in Hong Kong decreased by about 1.6 μg m^{-3}. The correlation coefficient at roadside MK station (0.70) is lower, likely because the emission variations near the single roadside station respond differently to the emission control programs compared to the greater Hong Kong region. The large intercept for the ordinary least squares regression (4.6 μg m^{-3}) may result from an underestimation of Hong Kong road transport emission inventory, non-local motor vehicle contributions to the resolved factor, or admixing of contributions from other largely carbonaceous source categories into the factor assigned to vehicle exhaust. These different scenarios do not alter the interpretation of the long-term trend but do affect the total PM_{10} SCE assigned to local vehicular exhaust and thus additional work is needed to further refine the motor vehicle contribution estimates.

In contrast to the decreasing trend for vehicle exhaust, annual average SCEs for residual oil increased from 1998 to 2004 and have been generally constant thereafter. In terms of relative changes the residual oil factor increased more than any other factor over the eleven year period. For the general stations, the residual oil factor annual average SCE was 0.3 μg m^{-3} in 2008 which is threefold higher than the contribution of 0.1 μg m^{-3} in 1998. The relatively high loading of sulfate onto this factor (Table S2) is not surprising. In Hong Kong, residual oil is mainly used as marine vessel fuel and oceangoing cargo vessels are legally allowed to use fuels with sulfur content up to 4.5%. In contrast, the sulfur content of motor diesel and petrol/gasoline dispensed in Hong Kong is currently capped at 0.005% (Gall and Van Rafelghem, 2006). While there are numerous marine vessel lanes throughout the navigable waters near Hong Kong and the greater PRD, the highest summertime residual oil combustion SCEs are observed at TW followed by SSP and MK (Fig. 3). This spatial pattern is consistent with significant emissions from the high density of container terminal operations located south and southwest of these three stations (Fig. 2).

Sea salt factors were classified as fresh or aged with the fresh being rich in chloride and the aged being depleted in chloride and enriched in sulfate and nitrate. Fig. 4 demonstrates that over the eleven-year period, fresh sea salt contributions to PM_{10} modestly decreased (2.0 μg m^{-3} for the five year period starting 1998 and 1.6 μg m^{-3} for the five year period starting 2004) while aged sea salt contributions increased (6.6 μg m^{-3} for the five year period starting 1998 and 7.4 μg m^{-3} for the five year period starting 2004). The annual average sodium ion concentrations for the general stations data were quite stable over the eleven-year period with a maximum deviation from the grand mean being 11%; this local maximum, observed in 2006, is reflected in the aged sea salt SCE time series. Similarly, sodium ion apportioned to the combined sea salt factors was stable over the study period with a maximum

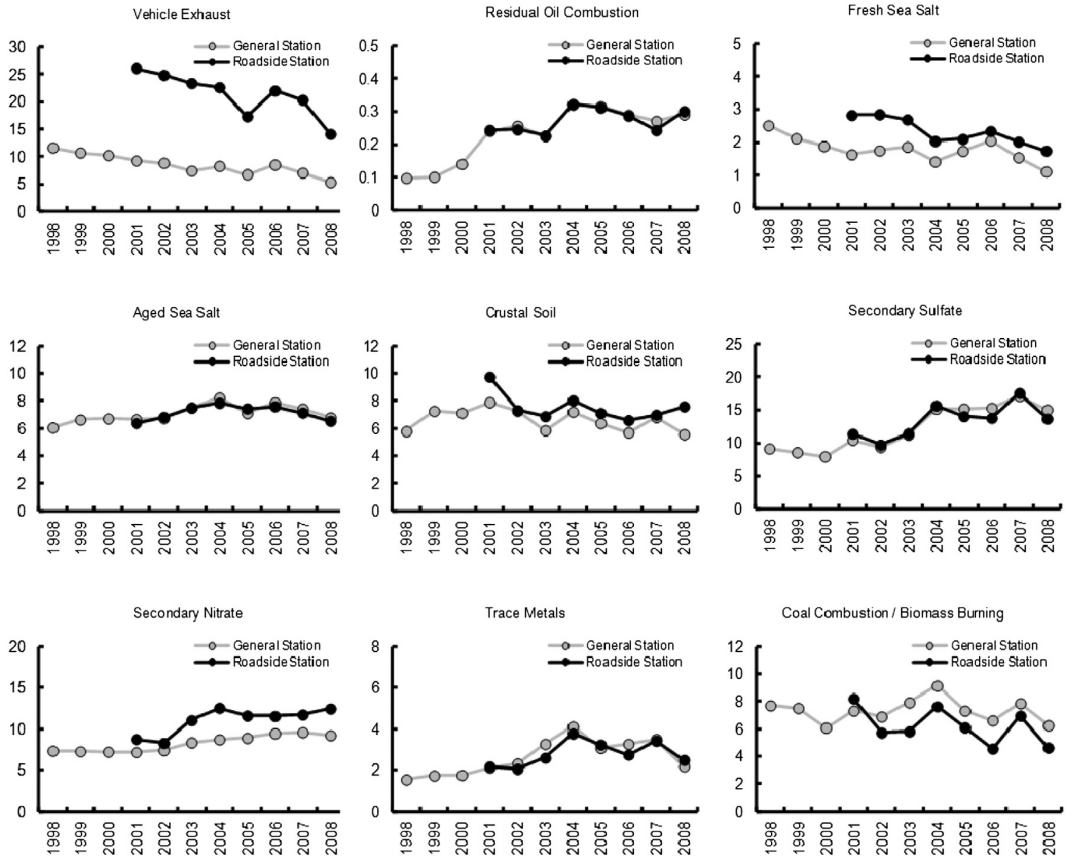

Fig. 4. PMF-resolved annual average source contribution estimates (unit: μg m^{-3}) at general stations and roadside station for the period 1998–2008.

annual deviation from the grand mean being 12%. Thus, the sea salt SCE patterns indicate negligible change in the sea salt emissions but an increase over time in the atmospheric processing of sea salt, perhaps in response to increased transport of inorganic PM from non-local sources as described below. This behavior drives an increase in overall sea salt SCEs because sulfate and nitrate have higher molecular weight than the chloride they are replacing.

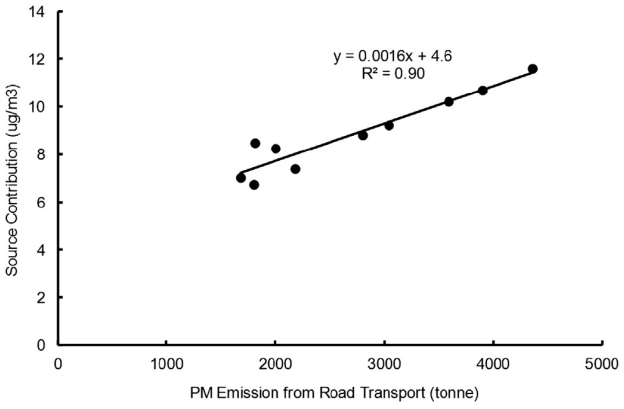

Fig. 5. Correlation between the PMF-resolved vehicle exhaust source contribution estimates at general stations and estimation of PM emissions from road transport. The emission inventory data are from HKEPD (2008). The data were fitted with an ordinary least squares regression.

Annual-average SCEs for the crustal soil/dust factor were relatively constant over the study period. Both spatial and temporal patterns suggest this factor has non-local origins. In addition to re-suspended material, this factor might have contributions from industrial sources. For example, a 2006 emission inventory for the PRD assigned more than half of the primary PM_{10} emissions to industrial sources with the dominant sub-categories being nonmetallic mineral products (He et al., 2011). This material might have composition similar to soil/dust.

Both secondary sulfate and secondary nitrate SCEs increased from 1998 to 2004 and have been relatively constant thereafter through 2008. Secondary sulfate SCEs for the general stations increased from 9 μg m^{-3} in 1998 to 17 μg m^{-3} in 2007, corresponding to a ~90% increase. Meanwhile, secondary nitrate SCEs at the general stations increased from 7 μg m^{-3} in 1998 to more than 9 μg m^{-3} in 2007, corresponding to a ~30% increase. Fig. 3 demonstrates that the spatial variability in sulfate is relatively small across the network with a modest gradient of decreasing SCEs from the north to the south. These spatial patterns and the large seasonal differences strongly implicate non-local sources as the dominant contributors to secondary sulfate in Hong Kong. Patterns for nitrate are generally similar to sulfate for the general stations; the behavior of nitrate at the MK roadside station is discussed in Section 3.3.

Contributions from the trace metals factor increased from 1998 to 2004 and have been relatively constant thereafter (Fig. 4). The local maxima in 2004 and 2007 track the secondary sulfate factor SCE and thus might result from inter-annual variability in synoptic transport patterns. This factor could represent a broad range of

industrial emission sources. A 2006 emission inventory for the PRD assigned 55% of the primary PM_{10} emissions to industrial sources with the dominant sub-categories being nonmetallic mineral products followed by pulp and paper industry and light manufacturing (He et al., 2011). It is possible that the PM_{10} mass loading onto the trace metals factor has substantial contributions from other industrial sources, such as those mentioned, that are located in the same region as the metals-emitting sources and are admixed into the factor.

There was no discernible trend for the SCEs of coal combustion/biomass burning factor over the period 1998–2008 with annual average contributions of 6.6 μg m^{-3} for the five year period starting 1998 and 6.5 μg m^{-3} for the five year period starting 2004. Similar to the secondary sulfate and trace metals factors, there were local maxima in 2004 and 2007 that might reflect differences in synoptic transport patterns. PRD biomass burning $PM_{2.5}$ emissions, which are dominated by domestic biofuel burning and field burning of rice straw, decreased by ~20% over the period 2003–2007 (He et al., 2011). However, a 2006 emission inventory assigned only 2% of the PRD primary PM_{10} emissions to biomass burning (Zheng et al., 2009b) and this change might be too small to detect in the ambient data. The same study allocated 21% of the PRD primary PM_{10} emissions to power plants. Emissions from industrial combustion sources could also be admixed into this factor.

Fig. 6 shows the long-term trend of relative contributions from non-local sources for winter, summer, transition seasons, and all sampling days. The contribution of non-local sources at the general stations increased from 58% in 1998 to 72% in 2004 (~7 μg m^{-3} increase) and has been relatively constant thereafter albeit with some year-to-year variability. This trend arises from the net effect of the decreasing contributions from vehicle emissions, driven by emission controls, being more than offset by the increase in contributions from non-local sources such as secondary sulfate, secondary nitrate, and trace metals. Percentage contributions from non-local sources are highest during the wintertime and increased from 72% in the winter of 1998–1999 to 86% in the winter of 2007–2008. In the latter case, contributions from local sources are estimated to be only 1/7 of the wintertime ambient PM_{10} and this demonstrates the increased importance of controlling sources from the PRD and beyond to improve air quality conditions in Hong Kong.

3.3. General station vs. roadside station

As expected, contributions from vehicle exhaust are much greater at the roadside stations compared to the general stations with grand average (1998–2008) vehicle exhaust SCEs of 8.5 μg m^{-3} for the general stations and 21.3 μg m^{-3} at the MK roadside station (Table 4). Fig. 4 and Table 4 demonstrate relatively similar SCEs between the general stations and the roadside station for all remaining emission source categories except fresh sea salt, secondary nitrate, and coal combustion/biomass burning. Elevated SCEs for fresh seas salt at MK results from its geographical location in the center of Kowloon Peninsula; the station is relatively close to the sea compared with those stations in the New Territories. Indeed, Fig. 3 shows that the fresh sea salt SCE at MK is quite similar to the SCEs at the nearby SSP and KT stations. Secondary nitrate is also elevated at the roadside station compared to the composited general stations, indicating that apart from the non-local contribution, local contributions cannot be neglected at roadside. The tall buildings and narrow roads in urban Hong Kong create a street canyon effect that traps vehicle emissions and can lead to elevated nitrate concentrations (Kim et al., 2012). The coal combustion/biomass burning SCE is lower at the roadside station compared to the general stations. In this case, there are significant SCE gradients across the network with higher SCEs to the north and lower SCEs to the south; the roadside station SCE is comparable to SCEs for the nearby SSP and KT stations. Thus, with the exception of vehicle exhaust and secondary nitrate, both local and non-local sources impact the general stations and roadside stations in similar ways despite differences in the sampling environment including the sampling height.

3.4. Source characteristics during high PM days

Synoptic weather conditions are an important driver in the occurrence of pollution episodes. They either stabilize the boundary layer that degrade air ventilation, or favor pollutant transport from high emission areas. Adverse health effects can arise from both chronic and acute exposures to PM (Wong et al., 2002). Thus, there is motivation to not only reduce average PM levels but also to "shave the peaks" off the highest PM concentration days. For example, in the United States these objectives are operationalized through having both annual and 24-h standards for $PM_{2.5}$. This issue motivates the evaluation of whether certain emission source categories have disproportionately high contributions on high PM days compared to average behavior.

In this study, we set the threshold value for high PM to be 100 μg m^{-3}, the value of World Health Organization (WHO)'s interim target-2 and the proposed air quality objective/index for 24-h PM_{10} in Hong Kong. The threshold criterion of 100 μg m^{-3} yields a total of 408 high PM sample-days at the seven sites from

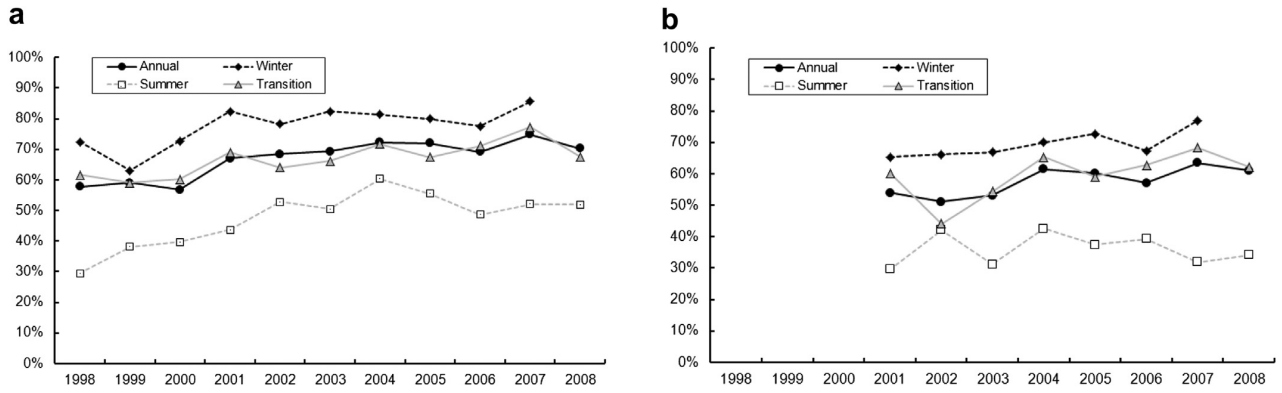

Fig. 6. Long-term trends of contribution percentages from non-local sources in winter, summer, transition season, and for all sampling days: (a) general stations; and (b) at roadside station.

Table 5
Year-specific number of high PM days (nominally 61 sample days per year). Asterisks denote years with significant data completeness gaps (Table 1).

No. high PM days	YL	TC	TW	SSP	CW	KT	MK
1998	11		4	4	3	2*	
1999	5	0*	7	2	3	3	
2000	7	5	3	5	2	1	
2001	8	3	2	1	3*	2*	10
2002	4	2	2	2	0*	4*	5
2003	8	8	4	3*	4	2	8
2004	15	5	8	1*	2	3	15
2005	15	3	7	7	9	6	11
2006	11	4	7	7	6	5	9
2007	10	10	7	3	6	8	17
2008	10	7	7	1	4	2	8
High PM days	104	47	58	36	42	38	83
Total days	654	575	631	586	641	598	482
% High PM days	15.9%	8.2%	9.2%	6.1%	6.6%	6.4%	17.2%

1998 to 2008. The number of days each year exceeding the PM_{10} mass measured at each station is listed in Table 5. For reference, a 1-in-6 day sampling schedule would have 61 sampling days per year for 100% data completeness. While the annual average PM_{10} mass has remained relatively constant over the eleven year study period (Fig. 1), the frequency of high PM days at the general stations nearly doubled for the five year period starting 2004 compared to the five year period starting 1998.

Fig. 7 shows the frequencies of high PM days (numerical values provided at the bottom of Table 5) and the average source contributions by the nine identified sources at individual stations on high PM days during 1998–2008. Apart from the roadside station, the northern most general station in the network, YL had the highest frequency of high PM days (16%). SSP, KT and CW, located in the southern portion of the HKSAR, had the lowest frequency of high PM days (6–7%). Overall, the frequency of high PM days in Hong Kong was 10% during 1998–2008.

The average mass concentrations on high PM days were similar among all stations (Fig. 7), with a variation of only 2%, despite the stations being distinct in their local environments (i.e., residential, commercial, industrial, roadside, or near container terminal). The highest average mass concentration (125 μg m^{-3}) occurred at YL and the lowest average mass concentration (118 μg m^{-3}) occurred at KT. Both the highest frequency and the highest mass concentration occurred at YL in the northwest, while both the lowest frequencies and lowest mass concentrations occurred at CW, TC and KT in the southeast. The narrow range of average

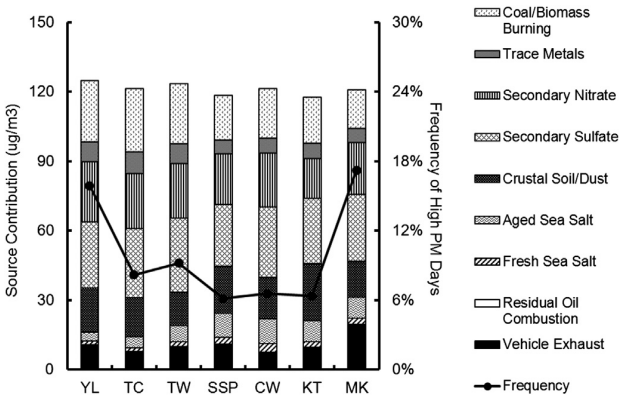

Fig. 7. Frequencies of the high PM days by site (i.e., the number high PM samples divided by the total number of collected samples at each station) and the PMF-resolved source contribution estimates averaged over the high PM days.

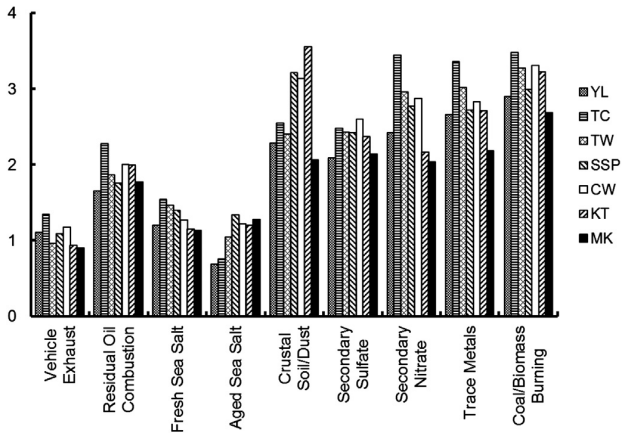

Fig. 8. Ratios of average source contribution estimates on high PM days to the source contribution estimates averaged over all samples.

concentrations on high PM days and the concentration gradient from northwest to southeast suggests that the non-local PM sources, especially in areas to the north of Hong Kong including the PRD, are significant contributors on high PM days.

The source contributions on high PM days were compared with the average values over the 1998–2008 study period to identify the sources that had elevated contributions on the high PM days. Fig. 8 plots the ratios of average source contributions on high PM days to that for the entire study period at individual stations. Mass contributions from the three local emissions source categories – vehicle exhaust, fresh and aged sea salt – did not increase significantly on high PM days. While the residual oil combustion (the remaining local source) contribution on high PM days was at most twice the study average, the absolute value itself is less than 1 μg m^{-3} and thus is not a major contributor to the total mass on high PM days. In contrast, contributions from crustal soil, secondary sulfate, secondary nitrate, trace metals and coal combustion/biomass burning were on average two to three times higher on high PM days. These observations indicate that pollutant transport from outside Hong Kong is disproportionately responsible for high PM days rather than accumulation of local pollutants due to unfavorable atmospheric dispersion over the region.

4. Conclusions and policy implications

PM_{10} chemical speciation network data for Hong Kong collected from 1998 to 2008 was used to apportion PM_{10} mass to emission source categories. Spatial and seasonal patterns in the PMF-resolved source contribution estimates were used to classify sources as local (emissions largely from within the HKSAR) or non-local (emissions largely from the PRD region and more distant sources). The time series of annual-average SCEs were examined for each factor. There is strong evidence for the effectiveness of local control measures on vehicle exhaust with contributions at the general stations decreasing more than 50% between 1998 and 2008. Most of this decrease occurred prior to 2004 which is consistent with the timeline for implementation of measures to reduce vehicle emissions (HKEPD, 2011). During this period, the PMF-modeled annual average source contribution estimates for vehicle exhaust decreased linearly with the HKEPD road transport PM emission inventory. Vehicle-related emission control strategies implemented over the previous fifteen years have resulted in a substantial positive impact by decreasing ambient burdens of – and thus

exposures to – vehicle exhaust. In contrast to the late 1990s, vehicle exhaust is no longer the dominant emission source category contributing to PM_{10} burdens at the general stations, with its contribution decreased from 22% in 1998 to 10% in 2008.

The PM_{10} air quality improvements from vehicle emissions controls have been offset by increased contributions from non-local sources which increased from being 58% of the PM_{10} burden in 1998 to 70% in 2008. In wintertime, the season with persistently high levels of PM air pollution, the relative contribution from non-local sources is even higher, increasing from 72% in the winter of 1998–1999 to 86% in the winter of 2007–2008. If current air quality conditions are maintained then on average the existing monitoring stations in Hong Kong will violate the proposed 24-h PM_{10} AQO of 100 $\mu g\ m^{-3}$ on about 10% of days. Future air quality conditions in Hong Kong will depend upon cooperation from provinces in mainland China. Such cooperation is already underway with Guangdong Province and perhaps a coordinated, regional-scale air quality management plan will best serve both the Hong Kong and neighboring provinces.

Source apportionment modeling suggests that PM from residual oil combustion has increased over the study period. These emissions are attributed to marine vessels and this increase is generally consistent with increased marine activity in the greater PRD region over the past decade. While the absolute PM mass impacts from this source category appear to be relatively small, residual oil combustion emissions do warrant further attention because these emissions tend to be rich in air toxics metals such as nickel (Lau et al., 2003). From a public health standpoint there is substantial motivation to better characterize marine-related activities/emissions and consider the implementation of air quality management plans targeting this sector such as near-shore fuel switching or more stringent fuel quality specifications. Such programs will require cooperation with Guangdong to be most effective.

Residual oil combustion is not the only source of air toxics metals in Hong Kong. For example, As, Cd, and Pb loaded onto the coal combustion/biomass burning factor and these same elements plus Mn loaded onto the trace metals factor. These metals can have several adverse effects such as acting as biochemical catalysts to cause severe molecular damage and induction of biochemical synthesis pathways (Halliwell and Gutteridge, 1999). Ambient levels of air toxics metals should be evaluated from a risk perspective to assess whether targeted controls are warranted.

The prominent role that emissions from sources outside the HKSAR exert on Hong Kong PM air quality highlights the need for regional air quality planning and management. Hong Kong and Guangdong have already taken a pioneering role in establishing China's first Regional Air Quality Monitoring Network. The monitoring results are released on the web as a Regional Air Quality Index (GDEPB, 2012). Since March 2012, Hong Kong and Guangdong has started releasing the hourly pollutant concentration data, including $PM_{2.5}$, to the public. This is the first city cluster in China to regularly release hourly pollutant concentration data. In recent years, a series of control measures were adopted in Guangdong to reduce pollutant emissions. Measures included requiring all large-scale thermal power plants to install and operate flue gas desulphurization units and to install continuous emissions monitoring systems with real-time on-line access by local authorities, closing down small power plants and other major polluting industries (e.g., cement plants and iron and steel plants with low production capacity), restricting the growth of the motorcycle populations in key cities, requiring newly registered motor vehicles in the PRD to comply with the National III standards (on a par with the Euro III standards), supplying the National III standard motor fuels, increasing use of nuclear power and natural gas, etc. These actions are laudable, yet additional measures will be needed to reverse the trend of deteriorating air quality in Hong Kong and the greater PRD region.

Although emissions from non-local sources have increased in relative importance, vehicle emissions remain among the most dominant source categories at the roadside station and further vehicle emission reductions within Hong Kong would be beneficial. For example, Lau et al. (2007) used SO_2 as a surrogate to classify daily air pollution burdens in 2006 as being dominated by local or regional sources. They estimated that Hong Kong was predominantly affected mainly by regional sources on 132 days (or 36% of the time) and by local sources on 192 days (53% of the time). The findings were significant, as it showed that people in Hong Kong were exposed to local pollution for longer periods of time over a year, hence measures that target local emissions would also greatly reduce public's exposure to air pollutants and the health risks associated with them. In addition, advances in the performance of internal combustion engines often results in more ultrafine particles which may have significant health impacts (Delfino et al., 2005). The European Union is now looking into the prospect of regulating ultrafine particles starting in 2013 (ECFA, 2011). These issues should be considered when planning the next round of vehicle emissions control and may motivate interest to convert the existing vehicle fleet to low- or no-emission models to comprehensively address the vehicle emissions problem.

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Appendix A. Supplementary information

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2012.09.026.

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