

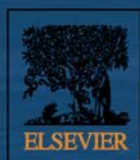
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Could wastewater analysis be a useful tool for China?



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Do vehicular emissions dominate the source of C6–C8 aromatics in the megacity Shanghai of eastern China?

Hongli Wang^{1,2}, Qian Wang², Jianmin Chen^{1,*}, Changhong Chen^{2,*}, Cheng Huang², Liping Qiao², Shengrong Lou², Jun Lu²

1. Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention (LAP³), Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China. E-mail: wanghl@saes.sh.cn

2. State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex, Shanghai Academy of Environmental Sciences, Shanghai 200233, China

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ABSTRACT

The characteristic ratios of volatile organic compounds (VOCs) to i-pentane, the indicator of vehicular emissions, were employed to apportion the vehicular and non-vehicular contributions to reactive species in urban Shanghai. Two kinds of tunnel experiments, one tunnel with more than 90% light duty gasoline vehicles and the other with more than 60% light duty diesel vehicles, were carried out to study the characteristic ratios of vehicle-related emissions from December 2009 to January 2010. Based on the experiments, the characteristic ratios of C6–C8 aromatics to i-pentane of vehicular emissions were 0.53 ± 0.08 (benzene), 0.70 ± 0.12 (toluene), 0.41 ± 0.09 (m,p-xylenes), 0.16 ± 0.04 (o-xylene), 0.023 ± 0.011 (styrene), and 0.15 ± 0.02 (ethylbenzene), respectively. The source apportionment results showed that around 23.3% of C6–C8 aromatics in urban Shanghai were from vehicular emissions, which meant that the non-vehicular emissions had more importance. These findings suggested that emission control of non-vehicular sources, i.e. industrial emissions, should also receive attention in addition to the control of vehicle-related emissions in Shanghai. The chemical removal of VOCs during the transport from emissions to the receptor site had a large impact on the apportionment results. Generally, the overestimation of vehicular contributions would occur when the VOC reaction rate constant with OH radicals (k_{OH}) was larger than that of the vehicular indicator, while for species with smaller k_{OH} than the vehicular indicator, the vehicular contribution would be underestimated by the method of characteristic ratios.

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Introduction

Volatile organic compounds (VOCs) are important precursors of the photochemical process and have received a large amount attention due to their great contribution to the formation of troposphere ozone and secondary organic aerosols (SOA) (Seinfeld and Pandis, 2006; IPCC, 2007). SOA from urban sources may be the dominant source of organic aerosols globally between 30° and 50° latitude (de

Gouw and Jimenez, 2009). The VOCs responsible for the majority of urban SOA formation have been assumed to be light aromatics with one benzene ring (Odum et al., 1997). According to estimates, global aromatics resulted in SOA production of 5.5 Tg/year (Liu et al., 2012), which contributed to around one third of the total SOA production (Volkamer et al., 2006). Furthermore, aromatics have been recognized as the most important contributors to the reactivity of VOCs and the ozone formation potential (OFP) in many megacities in

* Corresponding authors. E-mails : jmchen@fudan.edu.cn (Jianmin Chen), chench@saes.sh.cn (Changhong Chen).

eastern China (Zhang et al., 2007; Shao et al., 2011; Wang et al., 2013a). Total aromatics emissions over China were estimated at 13.4 Tg/year from the top-down inventory, mainly distributed in eastern China (Liu et al., 2012).

There are various kinds of emission sources of aromatics in the atmosphere, including vehicle exhaust, petrochemical and chemical industry, solvent use, coal burning, even biomass burning. However, few studies were focused on the source apportionment of aromatics up to now, although there have been a lot of studies on the source apportionment of the ambient VOCs in many cities and regions either by receptor models or by emission inventories (Huang et al., 2011; Yuan et al., 2012). Those source apportionment results of total VOCs have been helpful in enabling the government to control the major VOC emission sources. However, per unit decreases of VOCs from different sources might result in different improvements in air quality because of the differences of source profiles in emission sources. The air quality should be more sensitive to the decrease of sources with more reactive species. This suggests that the source apportionment of reactive species is essential.

Among many sources, vehicular emissions have been proved to be a very important contributor of VOCs in many megacities (Lau et al., 2010; Wang et al., 2010; Ling et al., 2011; Zhang et al., 2013). As reported, there have been a great number of vehicles, with the amount over three million until 2010 (<http://www.stats-sh.gov.cn/siteSearch/search.xhtml>) in Shanghai, which dominated VOC emissions and contributed over one third of total VOCs in the urban area (Cai et al., 2010; Wang et al., 2013a). One important question concerns the vehicular contribution to the reactive species. According to our previous study (Wang et al., 2013a) in Shanghai, aromatics accounted for 36% of the initial VOCs in the ambient, and more importantly C7–C8 aromatics dominated (more than 50%) the consumed VOC species, which agreed well with the oxidant production. To the best of our knowledge, there have been few studies on the vehicular contribution to the reactive species in Shanghai, in particular C7–C8 aromatics.

Given the facts mentioned above, this study aimed at assessment of vehicular and non-vehicular contributions to C7–C8 aromatics in urban Shanghai by using the characteristic ratios of these reactive species to the exclusive vehicular indicator, which were obtained through tunnel experiments. In addition, benzene, one relatively inert aromatic species, has aroused growing concerns due to its health risk to human beings (Cocheo et al., 2000; WHO, 2000; Zhang et al., 2012). Thus, the source analysis of benzene was also studied. The impact of chemical loss of VOCs during the transport from emission to the receptor site on the source analysis was taken into account.

1. Experiments

1.1. Tunnel experiments

VOC samples were collected in two kinds of typical tunnels in urban Shanghai from December 2009 to January 2010. One was Yan'an tunnel in the center of Shanghai city, and more than 90% of vehicles driving through this tunnel were light duty gasoline vehicles. The other tunnel was Dapu tunnel in the boundary of the urban area, and around 60% of vehicles driving in this tunnel were heavy duty diesel vehicles. The samples were collected in the vent of the tunnel using Summa canisters (Entech, USA) in two rush hours (8:30–9:30 and 17:00–18:00) and at noontime (12:00–13:00). In all, 16 samples (6 samples in Yan'an tunnel, and 10 samples in Dapu tunnel) were collected using Summa canisters in tunnel experiments. The composition of VOCs from the tunnels has been reported in our previous study (Lu et al., 2010).

1.2. VOCs sample analysis

VOC samples collected in tunnel were analyzed by a Gas Chromatograph Mass Spectrometer (GC-MS) system (GC7890A/MSD5975C, Agilent Technologies, USA). Samples were first concentrated by a cryogenic pre-concentrator (Entech 7100, USA), and then injected into the gas chromatograph and quantified using a quadrupole mass spectrometer. The PAMS (Photochemical Assessment Monitoring System) standard gas (Spectra, USA) was used to confirm compounds' retention time and identify compounds. In all, 56 species were identified and measured in this study. The method detection limits (MDLs) of the various VOC species ranged from less than one to several tens of micrograms per cubic meter (Qiao et al., 2012). A detailed description of the method can be found in our previous study (Wang et al., 2014).

Ambient C6–C8 aromatics, specifically benzene, toluene, xylenes (m-, p-, o-), styrene, and ethylbenzene and i-pentane, were continuously measured by an on-line gas chromatograph with flame ionization detector (GC-FID) system (Chromato-sud airmoVOC, France, Bordeaux) on the roof of a five-story building in urban Shanghai in 2010. The PAMS standard gas (Spectra, USA, Houston) was used to calibrate the instrument. Detailed description of the measurement can be found in the previous study (Wang et al., 2013a).

1.3. Estimation of VOC initial ratios

The initial ratios of VOCs were estimated based on the chemical loss of VOCs by reaction with OH radicals. The photochemical process of the air mass can generally be tracked through the evolution of the ratio between two hydrocarbons from similar emission sources but having different reactivities with OH. In this study, ethylbenzene (E) and m,p-xylenes (X) were selected for this purpose (Nelson and Quigley, 1983). The initial mixing ratio was calculated by Eqs. (1) and (2) (McKeen et al., 1996). More details of the calculation method of the initial ratios can be found in our previous study (Wang et al., 2013a).

$$[\text{VOC}_i]_t = [\text{VOC}_i]_0 \times \exp(-k_i[\text{OH}]\Delta t) \quad (1)$$

$$\Delta t = \frac{1}{[\text{OH}](k_E - k_X)} \times \left(\ln \left\{ \frac{[\text{E}]}{[\text{X}]} \right\}_{t=t_0} - \ln \left\{ \frac{[\text{E}]}{[\text{X}]} \right\}_{t=t} \right) \quad (2)$$

where, $[\text{VOC}_i]_t$ and $[\text{VOC}_i]_0$ are the observed and initial mixing ratio of VOC_i , respectively. k_i is the reaction rate constant of VOC_i with OH radicals. $[\text{OH}]$ is the mixing ratio of OH radicals. Δt is the photochemical age of the air mass. $([\text{E}]/[\text{X}])_{t=t_0}$ is the ratio between the initial mixing ratios of ethylbenzene and m,p-xylenes, and can be obtained by the ratio of [E] to [X] in the VOC emission sources. In this study, the $([\text{E}]/[\text{X}])_{t=t_0}$ was selected as 0.5, and the detailed description of the selection of the this ratio was discussed in the previous study (Wang et al., 2013a). $([\text{E}]/[\text{X}])_{t=t}$ is the ratio of [E] to [X] at time t .

The method above could result in very large uncertainties for the species with larger k_{OH} than m,p-xylenes (de Gouw et al., 2005). Thus, the initial mixing ratio of styrene, whose k_{OH} is larger than that of m,p-xylenes, was estimated in this study

using the rate coefficient of *m,p*-xylenes in Eq. (1) rather than its coefficient.

2. Results and discussion

2.1. Selection of the vehicular indicator

The ratios and correlations between specific VOC species pairs have been used in the identification of VOC emission sources (Gelencsér et al., 1997; Chan et al., 2002; Na, 2006; Hoshi et al., 2008; Chang et al., 2009; Wang et al., 2010; Yuan et al., 2012). In principle, compounds exclusive to vehicular emissions can serve as an indicator of vehicular emissions and a basis for calculating the relative abundance of other species. Then, the contribution of the non-vehicular and vehicular sources can be determined for an area based on the characteristic ratio of the indicator to other species (Borbon et al., 2001; Chang et al., 2006, 2009). The basic criterion for being a vehicular indicator is that its atmospheric presence is solely and exclusively from vehicular sources. Thus, selecting one suitable species to be used as the indicator of vehicular emissions is of great importance.

In the previous studies, some species were employed as vehicular tracers, such as acetylene, benzene, 3MC5A (3-methylpentane), 3MC6A (3-methylhexane), and *i*-pentane (Na et al., 2001; Chang et al., 2003, 2009). However, in the regions with industrial emissions, acetylene or benzene may not be suitable as a vehicular indicator. As reported by Chang et al. (2006), 3MC5A, 3MC6A (methylcyclopentane), 2MC6A (2-methylhexane) and 3MC6A were mostly (about 75%–100%) emitted from motor vehicles by both tailpipe exhaust and gasoline evaporation in Taiwan, and thus could be used as the tracers of vehicular emissions in Taiwan.

Recently, *i*-pentane has been reported as one of the top three most abundant components in the tunnel of Taiwan along with ethylene and acetylene (Lai and Peng, 2011). According to the profiles of major VOC sources in the megacities of China, *i*-pentane was also a very considerable VOC species (Liu et al., 2008a; Wang et al., 2013b) in the emissions of vehicles. Specifically, *i*-pentane accounted for ca. 8% of VOC emissions both from gasoline vehicle exhaust and evaporation and ca. 4% of VOC emissions both from diesel vehicle exhaust and evaporation (Liu et al., 2008a). Based on the measurements in this study as shown in Table 1, *i*-pentane contributed 5.1%–8.2% of VOCs with an average contribution of 6.2%. By contrast, the contribution of *i*-pentane to VOC emissions from other major sources, including asphalt, industrial and residential coal burning, biomass burning, and the petrochemical industry,

was negligible, with proportions lower than 1% (Liu et al., 2008a). Wang et al. (2014) reported that *i*-pentane made up 0.2% of VOC emissions from solvent use in Shanghai. Besides the source characterization of VOC emissions as mentioned above, existing publications also supported that more than 90% of *i*-pentane was from vehicular emissions (including vehicle exhaust and fuel evaporation) even in Guangzhou, where variable and complex industrial sources are distributed (Liu et al., 2008b). Accordingly, *i*-pentane was employed as a suitable indicator of vehicular emissions. In addition, the mixing ratio of *i*-pentane was considerable in urban Shanghai, which allowed accurate and precise measurement (Wang et al., 2013a), and its lifetime in the atmosphere is relatively long (Atkinson et al., 2006).

2.2. Vehicular characteristic ratios of VOC species to *i*-pentane

When employing vehicular indicators to assess the vehicular contribution to VOC species in the ambient atmospheric environment, the vehicular characteristic ratios of VOC species to the indicator must be established first. Generally, these vehicular characteristic ratios vary with the vehicles and fuel types in different regions (Blake and Rowland, 1995; Liu et al., 2008a; Chang et al., 2009). Therefore, the representativeness of the vehicular characteristic ratio is very crucial. In this study, the measured ratios in tunnel were used for this purpose instead of those in vehicle exhaust, because VOC emissions in the tunnel were mixtures of VOCs from the various types of vehicles, and included both exhaust and fuel evaporation. Considering the differences in VOC profiles between gasoline vehicles and diesel vehicles (Liu et al., 2008a), two kinds of tunnels, namely a tunnel with more than 90% light duty gasoline vehicles and a tunnel with around 60% heavy duty diesel vehicles, were employed to study the vehicular characteristic ratios of VOC species to *i*-pentane.

The concentrations of C6–C8 aromatics and *i*-pentane measured in tunnel are listed in Table 1. The concentrations of these VOC species in Yan'an tunnel were much higher than those in Dapu tunnel, as discussed in our previous study (Lu et al., 2010). Interestingly, the ratios of C6–C8 aromatics to *i*-pentane in these two types of tunnels showed moderate agreement with each other, as shown in Table 2, with slightly higher ratios in Yan'an tunnel, characterized by gasoline vehicles, compared with those in Dapu tunnels, characterized by diesel vehicles emissions. As a result, average ratios of C6–C8 aromatics to *i*-pentane in the tunnel were obtained, namely 0.53 ± 0.08 (benzene), 0.70 ± 0.12 (toluene), 0.41 ± 0.09 (*m,p*-xylenes), 0.16 ± 0.04 (*o*-xylene), 0.023 ± 0.011 (styrene), and 0.15 ± 0.02 (ethylbenzene), respectively, which were used to estimate the vehicular contributions of C6–C8 aromatics in this study. Our results for the ratios between *i*-pentane and C6–C8 aromatics were higher than those in Guangzhou (Liu et al., 2008a) to some extent, as shown in Table 2, possibly due to differences in the vehicles in Shanghai and Guangzhou.

2.3. Vehicular and non-vehicular contributions

Generally, the relative contributions of vehicular and non-vehicular sources to VOCs in the atmosphere could be

Table 1 – Concentrations of VOCs measured in tunnel of urban Shanghai (unit: ppbv).

VOC _i	Mean	Range
<i>i</i> -Pentane	36.93 ± 16.64	20.32–67.66
Benzene	20.50 ± 11.44	8.60–42.29
Toluene	26.91 ± 15.45	12.06–58.10
<i>m,p</i> -Xylenes	16.20 ± 10.15	5.11–36.69
<i>o</i> -Xylene	6.42 ± 3.95	1.94–14.41
Styrene	0.90 ± 0.49	0.08–1.85
Ethylbenzene	5.98 ± 3.54	2.85–13.44
VOCs	634.9 ± 347.7	247.5–1275.1

Table 2 – Ratios (mean \pm standard deviation) of C6–C8 aromatics to i-pentane in tunnel and in ambient.

VOC _i /i-pentane	Yan'an tunnel	Dapu tunnel	Liu et al. (2008a)	Observed ^a	Initial ^b
Benzene/i-pentane	0.62 \pm 0.03	0.48 \pm 0.03	0.41	0.95 \pm 0.41	0.86 \pm 0.37
Toluene/i-pentane	0.83 \pm 0.08	0.62 \pm 0.04	0.56	2.82 \pm 1.11	3.12 \pm 1.25
m,p-Xylenes/i-pentane	0.51 \pm 0.04	0.35 \pm 0.06	0.41	1.14 \pm 0.40	2.36 \pm 1.01
o-Xylene/i-pentane	0.20 \pm 0.02	0.14 \pm 0.02	0.14	0.45 \pm 0.15	0.72 \pm 0.27
Styrene/i-pentane	0.03 \pm 0.01	0.02 \pm 0.01	0.03	0.12 \pm 0.02	0.23 \pm 0.14
Ethylbenzene/i-pentane	0.18 \pm 0.02	0.14 \pm 0.01	0.14	0.99 \pm 0.40	1.18 \pm 0.50

^a Ratio based on the observed mixing ratio in ambient.

^b Ratio based on the initial mixing ratio in ambient; Liu et al. (2008a), the tunnel results in Guangzhou.

apportioned after the vehicular characteristic ratios of VOC species to indicator were obtained. However, the chemical loss of VOCs during the transport from emissions to the monitoring sites could influence the ratio of each VOC species to the indicator. Consequently, the apportionment of vehicular and non-vehicular contributions to VOCs would be inaccurate if this chemical removal were neglected. Therefore, in this study, the initial mixing ratios of VOCs were estimated by the methodology described in Section 2.3, and the ratio between the initial mixing ratios of VOC species was employed to estimate the contribution of vehicular emissions, combined with the vehicular characteristic ratio measured in tunnel.

Fig. 1 shows the correlations of C6–C8 aromatics to i-pentane measured in tunnel, and also the initial mixing ratios of these species in ambient air are plotted against that of i-pentane. The points lying on the regression line mean that the samples are exclusively from vehicular emission, while the points lying above the regression line suggest that these samples are both from vehicular and non-vehicular emissions. Based on the average ratio between the initial mixing ratios of VOC species and i-pentane, the relative contributions from vehicular and non-vehicular emissions could be estimated, combined with the vehicular characteristic ratio shown in Table 2. For comparison, the observed mixing ratios of the species in the ambient are also plotted against that of i-pentane in Fig. 1. The apportionment results are summarized in Table 3.

2.3.1. Vehicular and non-vehicular contributions

The average ratio between the benzene initial mixing ratio and that of i-pentane was 0.86 ± 0.37 , while this ratio was 0.53 ± 0.08 in tunnel. Therefore, the relative contribution of vehicular emission to benzene was 61.6%; while the average ratio between observed benzene and i-pentane was 0.95 ± 0.41 , and the vehicular contribution was 55.7%, lower than that based on the initial ratio.

For toluene, the average ratio between its initial mixing ratio and i-pentane was 3.12 ± 1.25 . This ratio in tunnel was 0.70 ± 0.12 . Hence, the vehicular emission amounted to 22.4% for toluene. Obviously, the non-vehicular emission was more important for the source of toluene than for benzene. In terms of the ratio between the observed mixing ratios, the value was 2.82 ± 1.11 , and the vehicular emission derived from this value was 24.8%. The contribution from vehicles was slightly overestimated based on the ratio between the observed mixing ratios.

In the case of m,p-xylenes, the average ratio of its initial mixing ratio to that of i-pentane was 2.36 ± 1.01 . This ratio in the tunnel was 0.41 ± 0.09 . Thus, 17.4% of m,p-xylenes were derived from vehicular emission. The ratio between the observed mixing ratios was 1.14 ± 0.40 , and thus the vehicle contribution was 36.0%. Clearly, the vehicle contribution to m,p-xylenes is overestimated by over 100% derived from the observed mixing ratios. The result of o-xylene was similar to that of m,p-xylenes. The vehicular emission accounted for 22.2% of o-xylene, much lower than the value of 35.5% based on the observed ratios.

The average ratio of styrene initial mixing ratio to that of i-pentane was 0.23 ± 0.14 , compared to 0.023 ± 0.011 in the tunnel, and hence the vehicular contribution to styrene was 8.7%; while the result based on the observed data was 17.3%, twice as high as that derived from the initial data.

Similarly, the vehicular contribution to ethylbenzene was estimated as 12.7% according to the ratios of ethylbenzene to i-pentane in ambient and in tunnel environments. The vehicular contribution increased to 15.2% for ethylbenzene if the chemical removal was neglected.

On the basis of vehicular contributions to the aromatic species mentioned above, it could be concluded that around 23.3% of C6–C8 aromatics in urban Shanghai were from vehicular emissions, combining their ambient concentrations as listed in Table 3. Huang et al. (2011) reported that the contribution from vehicular emissions accounted for about 15%–20% for the source of aromatics in the Yangtze River Delta region based on the inventory method. Our result was a little higher than that of Huang et al. (2011), probably due to the greater amount of vehicular emissions in the urban area. Wang et al. (2013a) reported that the vehicular emissions contributed to around one third of total VOCs in urban Shanghai. Our results suggested that non-vehicular emissions had more contribution of reactive species in urban Shanghai. Different from the other megacities of China, such as Beijing and Guangzhou (Liu et al., 2008b; Wang et al., 2010; Zhang et al., 2013), the non-vehicular emissions were more important than the vehicular emissions for the reactive VOC species in urban Shanghai, although there was a great amount of VOCs emitted from the heavy traffic.

2.3.2. Diurnal variations of the vehicular contribution to aromatics

Based on the method above, the hourly vehicular contribution to each aromatic species was calculated. Fig. 2 shows the diurnal variations of the vehicular contribution to C6–C8

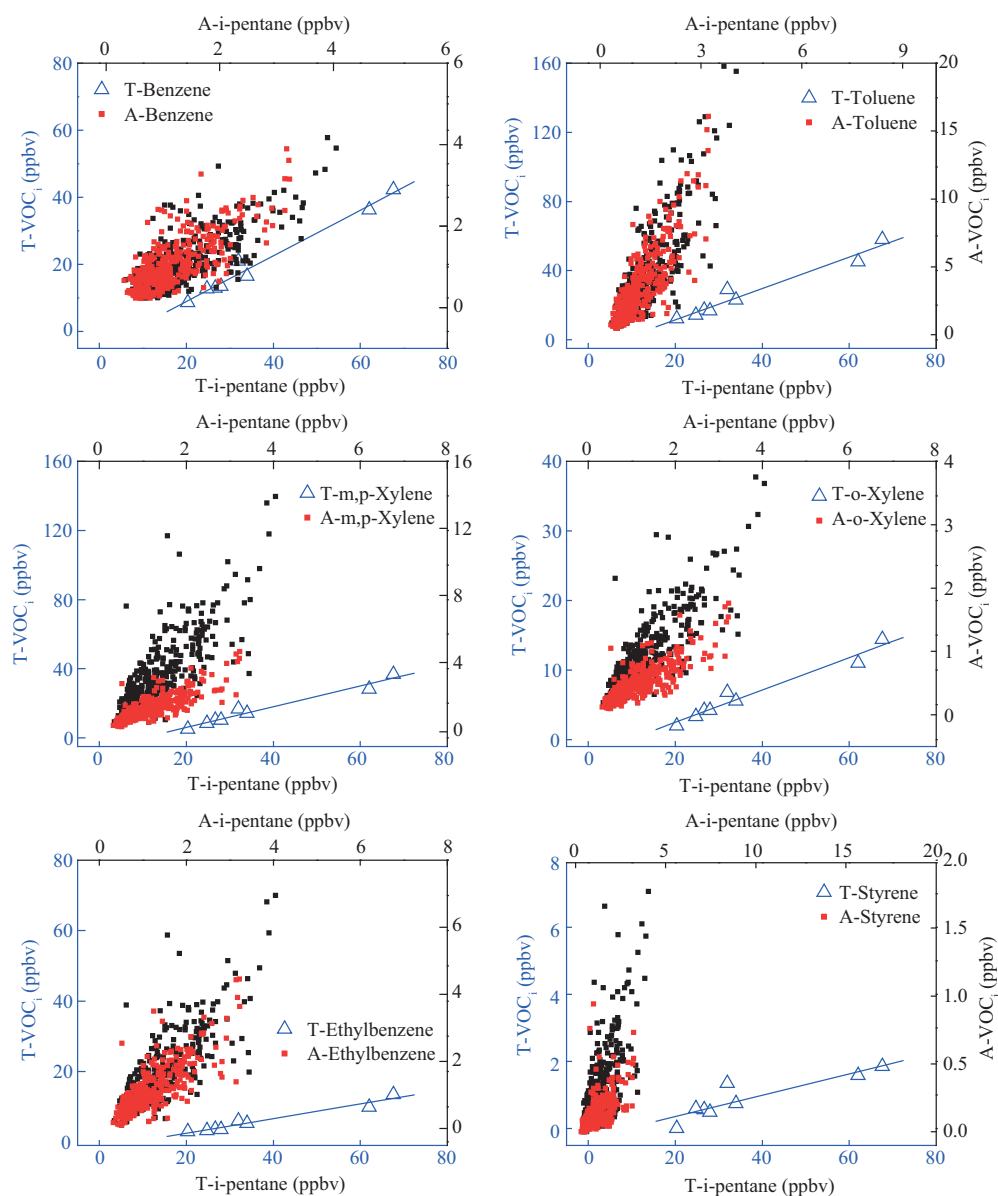


Fig. 1 – Scatter plots of the VOC_i species mixing ratio (average time: 8:00–18:00) to that of i-pentane in 2010 shown as solid squares (red: measured mixing ratio; black: initial mixing ratio) on the top and right axis; regression of the mixing ratios measured in the tunnel between VOC_i species and i-pentane, shown as open triangles on the left and bottom axis. (T is tunnel sample, and A is ambient sample.)

aromatics and the vehicle density of one typical major road in urban Shanghai that was about 8 km away from the monitoring site. The vehicle density was the average of the vehicle densities for ten successive days, including both weekdays and weekends. We can see that the vehicular contribution to C6–C8 aromatics estimated by the method above agreed well with the vehicle density. Specifically, the vehicular contribution decreased from ~30% during the rush hour in the morning to 22% at 11:00, due to the decrease of the vehicle density as shown in Fig. 2. The vehicular contribution showed a small peak during noon time, which could be explained by the peak on the diurnal pattern of vehicle density at the

corresponding time. The contribution of vehicles began to rise at 17:00 as the rush hour of evening started.

2.3.3. Uncertainty analysis

As mentioned above, the vehicular contribution for C7–C8 aromatics would be overestimated to different extents dependent on their chemical reactivities but would be underestimated for benzene, if the observed mixing ratios were directly used to identify the vehicular contribution. Fig. 3 shows the positive correlations of k_{OH} (reaction rate constant of VOC with the OH radical) with the overestimation percentage of the vehicular contribution based on the observed data. Accordingly, species

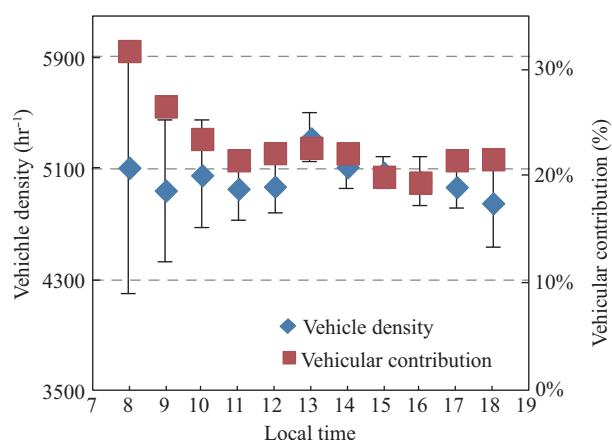
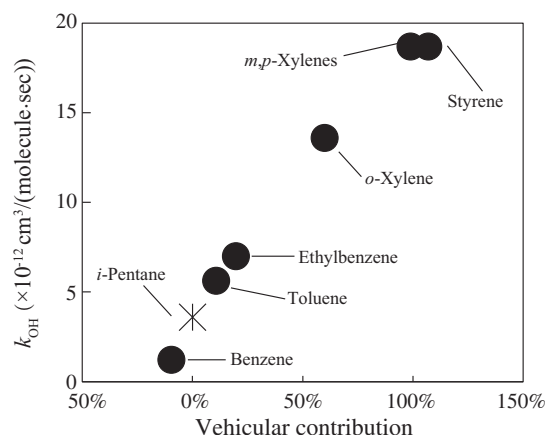
Table 3 – Concentration (mean \pm standard deviation) and vehicular contribution of C6–C8 aromatics in urban Shanghai.

VOC _i	Concentration, ppbv		Vehicular contribution	
	Observed	Initial	Observed ^a	Initial ^b
Benzene	1.07 \pm 0.63	1.13 \pm 0.67	55.7%	61.6%
Toluene	3.41 \pm 2.47	4.49 \pm 3.36	24.8%	22.4%
m,p-Xylenes	1.3 \pm 0.73	3.26 \pm 2.15	36.0%	17.4%
o-Xylene	0.51 \pm 0.27	0.98 \pm 0.59	35.5%	22.2%
Styrene	0.14 \pm 0.12	0.33 \pm 0.28	17.3%	8.7%
Ethylbenzene	1.14 \pm 0.70	1.63 \pm 1.08	15.2%	12.7%
Sum (C6–C8)	7.57	11.82	30.5%	23.2%

^a Based on the observed mixing ratio in ambient.^b Based on the initial mixing ratio in ambient.

with larger k_{OH} were usually consumed faster, which resulted in a larger overestimation of their vehicular contribution. However, the overestimation only occurred when the VOC k_{OH} was larger than that of the vehicular indicator species, namely *i*-pentane in this study. If the VOC k_{OH} was smaller than that of vehicular indicator, the vehicular contribution would be underestimated. As mentioned in Section 2.3, the initial mixing ratio of styrene was estimated using *m,p*-xylenes k_{OH} in Eq. (1), and thus similar results for styrene and *m,p*-xylenes were obtained in Fig. 3.

The uncertainties in calculating the contribution of vehicular and non-vehicular emissions by ratios based on the initial mixing ratio of VOCs mainly come from the estimation of the initial mixing ratio by $[E]/[X]_{t=t_0}$ and the measurement errors of the vehicular characteristic ratio of the indicator to VOC species in tunnel. Higher (0.7) and lower (0.3) $[E]/[X]_{t=t_0}$ were used to carry out the sensitivity analysis. The higher and lower $([E]/[X])_{t=t_0}$ values were obtained according to $[E]/[X]_{t=t_0}$ from many sources (Liu et al., 2008a; Wang et al., 2014). As a result, the relative changes of vehicular contribution to the C6–C8 aromatics ranged from 8% to 48% and from –6% to –54% for lower and higher $[E]/[X]_{t=t_0}$, respectively. Generally, the initial mixing ratios of reactive VOC species were more sensitive to $[E]/[X]_{t=t_0}$, and consequently the uncertainties of the vehicular contribution to reactive species were relatively high. The uncertainties of

**Fig. 2 – Diurnal patterns of the vehicle density and the vehicular contribution to C6–C8 aromatics.****Fig. 3 – Correlation of k_{OH} with the overestimation percentage of vehicular contribution based on the observed data.**

the measured vehicular characteristic ratios were from 13.3% to 25.0% in this study.

3. Conclusions

i-Pentane, as an important emitted species from vehicle exhaust and gasoline evaporation, was employed as the indicator of vehicular emissions because there was little emission of *i*-pentane from other sources in the megacities of China. The characteristic ratios of C6–C8 aromatics to *i*-pentane of vehicular emissions were measured in two kinds of typical tunnels in Shanghai, and were used to apportion the vehicular and non-vehicular contributions of C6–C8 aromatics.

The chemical removal of VOCs during transport had a large impact on the apportionment of vehicular and non-vehicular contributions to VOCs. Generally, overestimation of vehicular contribution would occur when the VOC k_{OH} is larger than that of vehicular indicator, while for species with smaller k_{OH} than that of vehicular indicator, the vehicular contribution would be underestimated by the method of characteristic ratios. Thus, the initial mixing ratio of VOCs was estimated according to the photochemical age of the air mass.

Based on the result, the characteristic ratios of C6–C8 aromatics to *i*-pentane of vehicular emissions were 0.53 ± 0.08 (benzene), 0.70 ± 0.12 (toluene), 0.41 ± 0.09 (*m,p*-xylenes), 0.16 ± 0.04 (*o*-xylene), 0.023 ± 0.011 (styrene), and 0.15 ± 0.02 (ethylbenzene), respectively in Shanghai. The apportionment result showed that around 23.3% of C6–C8 aromatics in urban Shanghai were from vehicular emissions, which meant that non-vehicular contributions were more important. These findings suggested that the emission control of non-vehicular sources, i.e. industrial emissions, should also receive attention in addition to the control of vehicle-related emissions in Shanghai.

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