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# Intra-Urban Levels, Spatial Variability, Possible Sources and Health Risks of $PM_{2.5}$ Bound Phthalate Esters in Xi'an

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## ABSTRACT

Phthalate esters (PAEs) are abundant semi-volatile organic compounds in fine particulate. $PM_{2.5}$ bound PAEs can inhale into the body with breath, which can cause negative effects to human health. In this study, total of 266 $PM_{2.5}$ samples dispersed from nineteen communities in Xi'an, were collected at December, 2013, the heavy pollution periods. Most of them are from residential areas, and four of them are in universities. Much high levels of PAEs were obtained in this study, which were from 271.7 to 2134 ng $m^{-3}$ (952.6 ng $m^{-3}$ on average). DEHP was the dominant species, with an average of 402.4 ng $m^{-3}$, and attributed for 42.2% of the total PAEs, followed by BBZP (146.8 ng $m^{-3}$ on average) and accounted for 15.4% of the total PAEs. Relative humidity and ventilation coefficient are the two meteorological factors affect the PAEs pollutions during the sampling periods. PAEs showed a declined trend from the urban to suburban. The principal component analysis (PCA) investigated that the release from plasticizer using in vinyl flooring, inks, synthetic leather, adhesives, and food contact wrapping; and emissions from cosmetics and personal care products, varnish, and volatilization from solid waste landfill or sewage sludge from wastewater treatment plant are the main sources for PAEs (86.8% of total PAEs). The daily inhalation and cancer risk assessment displayed that possible risk for all age group persons in this area, and infants are the most susceptible population.

***Keywords*:** Phthalate esters; Nineteen communities; Spatial variability; Possible sources; Health risks.

## INTRODUCTION

Phthalate esters (PAEs), as an organic chemicals raw materials, are widely used as plasticizers in various products, such as building materials, children's toys, medical devices, car products, clothing, and food packaging. They are also applied in adhesives, paint, and ink in home furnishings; and added to personal care products as fragrance (DEHA, 2000; Schettler, 2006; Guo and Kannan, 2013; Škrbic *et al.*, 2016; Yao *et al.*, 2016). Because PAEs are not chemical bounded to products, they are easily released into the environment during its manufacture, usage and disposal. They have been widely detected in various environmental matrixes, such as air, water, soil, sediment, road dust and so on (Zeng *et al.*, 2008; Song *et al.*, 2015; Net *et al.*, 2015; Al-Saleh *et al.*, 2017; Bamai *et al.*, 2016; Li *et al.*, 2016a; Li *et al.*, 2016b). Especially in the indoor environment (indoor air, indoor total suspend particulate, and house dust), PAEs are well documented in worldwide (Otake *et al.*, 2004; Rakkestad *et al.*, 2007; Gao and Wen, 2016; Škrbic *et al.*, 2016), for their abundant occurrence and with the long time stayed for humans in indoor environments (80–90%) (Castro *et al.*, 2011; Wang *et al.*, 2017a), which can cause serious potential exposure risks.

Toxicological researches investigated that some of PAEs can affect the growth of the male reproductive system

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(Adibi *et al.*, 2003; Axelsson *et al.*, 2015; Bornehag *et al.*, 2015; Bu *et al.*, 2016) and it also has correlations with the allergic symptoms and can increases the risk of asthma and bronchial obstruction for childhood (Ait Barmai *et al.*, 2014). Besides, the International Agency for Research on Cancer (IARC) (IARC, 1982) has listed DEHP as a possible carcinogen to humans (group 2B). Like most other organic toxic substances, PAEs can cause damage to human body through the inhalation, ingestion and dermal pathways (Bu *et al.*, 2016) and PAEs metabolites have been reported in breast milk, serum, and urine (Hogberg *et al.*, 2008; Guo *et al.*, 2011a; Gao *et al.*, 2016). Study has showed that their metabolites might more toxic to human body and even more some PAEs and their metabolites can cross the placenta barrier to potentially affect the fetal growth and development in early life (Preuss *et al.*, 2005; Wittassek *et al.*, 2009). And the children exposure to PAEs has been cause wide concern in the past years (Kasper-sonnenberg *et al.*, 2014; Kim *et al.*, 2014; Bao *et al.*, 2015).

China is one of countries that with the largest production and usage of PAEs (He *et al.*, 2013; Net *et al.*, 2015), and DEHP accounted for about 80% of the phthalates products (Meng *et al.*, 2014). Much high levels of PAEs were obtained in indoor air from hospital, new decorated house, indoor window films, besides the agricultural soil with plastic film mulching in Shandong Peninsula (Pei *et al.*, 2013; Wang *et al.*, 2015; Huo *et al.*, 2016; Li *et al.*, 2016c). The compositions of PAEs detected in these matrixes are close to the emission sources, and are very useful for the PAEs source apportionment. Recently years, with the heavy haze attacked most regions in China, the fine particulate matter ($PM_{2.5}$) cause widespread concern (Chen *et al.*, 2017; Zhang *et al.*, 2017). PAEs were abundant organic species in $PM_{2.5}$ (Wang *et al.*, 2006), and it can go into human body to cause negative health effects with the breath. Few researches were study on the concentrations and spatial distribution of PAEs in ambient air in China (Wang *et al.*, 2006; Li and Wang, 2015). The aims of this work are to determine inter-urban variations of PAEs in Xi'an in wintertime; to investigate the possible sources of PAEs and in addition, to estimate the inhalation risks of particulate bound PAEs.

## MATERIALS AND METHODS

### *Sampling*

Xi'an, the capital city of Shaanxi Province, has suffered severe air pollution in China for its geographic position, and it was always ranked within the last ten cities in China. Here filter samples were simultaneously collected at nineteen different communities dispersed in Xi'an (Cao *et al.*, 2005; Gao *et al.*, 2015; Wang *et al.*, 2016a) (Fig. 1). The details of the sampling environment and setup height of the equipment were described in Supplemental Table S1 (Wang *et al.*, 2016a). $PM_{2.5}$ samples were collected by Mini-Volume sampler (Airmetrics, Springfield, OR, USA) from 08:30 to 08:30 next day at a flow rate of 5 L $min^{-1}$, and loading on 47 mm quartz filters (QM/A®, Whatman Inc., U.K.). It was started from 2nd December to 15th December 2013. Blank samples were simultaneously collected at each sampling site at the 11th December. Total of 266 aerosol-loaded samples and 19 field blanks were collected in this study and all of the samples were properly stored in a freezer at –20°C before analysis. The origin of the meteorological parameters including temperature (T) and relative humidity (H), wind speed (U), and boundary layer height (MLH) were displayed in Supplemental materials and have been described in the previous study (Wang *et al.*, 2016a).

### *Chemical Analyses*

An in-injection port thermal desorption (TD) coupled with gas chromatography/mass spectrometry (GC/MS) was

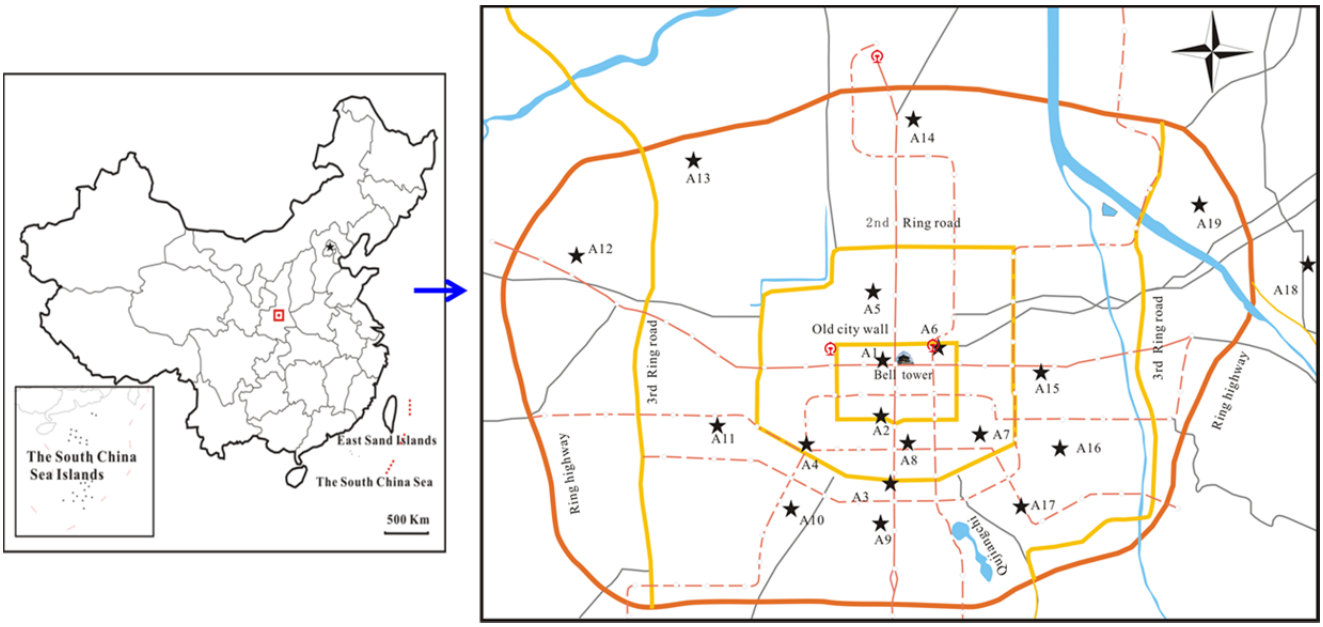


**Fig. 1.** Map of the sampling sites at Xi'an.

using for the analysis of $PM_{2.5}$ bound PAEs species, including dimethylphthalate (DMP), diethylphthalate (DEP), di-n-butyl phthalate (DBP), benzyl butyl phthalate (BBZP), bis (2-ethyl(hexyl))phthalate (DEHP), di-n-octyl phthalate (DNOP), and bis(2-ethylhexyl)adipate (DEHA) (Ho *et al.*, 2011; Wang *et al.*, 2016b). The detailed analyze process can be achieved in our previous studies (Ho *et al.*, 2008, 2011; Wang *et al.*, 2015; Wang *et al.*, 2017b).

### *Health Evaluation Model*

Inhalation is an important route for human exposure of $PM_{2.5}$ bound chemicals. Though the gas phase was not monitored here, the average daily inhalation dose of PAEs (EI, ng $kg^{-1}$ $d^{-1}$) by particle was also calculated by Eq. (1) and five age categories were classified: infants (< 1 year), toddlers (1–3 year), children (4–10 years), teenagers (11–18 years), and adults (≥ 18 years) (EPA, 1997; Guo and Kannan, 2011).

$$EI = \frac{C \times IR \times CF}{BW} \quad (1)$$

where C is the concentrations of the target pollutants in $PM_{2.5}$ (ng $m^{-3}$); IR is the inhalation rate ($m^3$ $day^{-1}$), it was 4.5, 7.6, 10.9, 14.0, 13.3 $m^3$ $day^{-1}$ for infants, toddlers, children, teenagers, and adults. CF is the exposure factor, 0.9; and BW is the body weight (kg), which was 5, 19, 29, 53, and 63 kg for infants, toddlers, children, teenagers, and adults (Zhang *et al.*, 2014).

For DEHP has a strong carcinogenic risk at low concentrations, the carcinogenic risk (CR) for PAEs, assessed by DEHP, and is identified by the International Agency for Research on Cancer (IARC) (IARC, 1982; Pei *et al.*, 2013; Li *et al.*, 2016b).

$$CR = q \times EI \times 10^{-6} \quad (2)$$

where q is the 95% upper confidence limit of the linearized carcinogenic slope factor for DEHP, that was 0.014 (mg $kg^{-1}$ $d^{-1})^{-1}$) (EPA, 1997). EI is daily inhalation dose (ng $kg^{-1}$ $d^{-1}$).

### *Quality Assurance and Control (QA/QC)*

The aerosol sampler was checked regularly and calibrated using a Defender 510 Volumetric Primary Flow Standard (Bios international Corporation, Butler, NJ, USA) during sampling. For instrument analysis, experiment and field blanks were analyzed simultaneously with the samples for quality control. Internal standard (IS) was added in each samples which contains chrysene-$d_{12}$ ($C_{18}D_{12}$) (98%, Sigma-Aldrich, Bellefonte, PA, USA), *n*-tetracosane-$d_{50}$ ($C_{24}H_{50}$) and phenanthrene-$d_{10}$ ($C_{14}D_{10}$) (98%, Aldrich, Milwaukee, WI, USA). Concentrations of PAEs were calculated with a five-point calibration over a concentration range of 40–400 ng. The relative standard deviation of replicate analysis (each ten samples) was from 0.5 to 8.2% for PAEs. Reported data were corrected for the average value of the blanks.

## RESULTS AND DISCUSSION

### *Concentrations of PAEs and the Meteorological Conditions Effects*

The concentrations of PAEs varied with the sampling sites (Fig. 2 and Supplemental Table S2). Each of 7 PAEs was detected in all filter samples with a range of 23.6 to 1482 ng $m^{-3}$ and total of PAEs were from 271.7 to 2134 ng $m^{-3}$ (952.6 ng $m^{-3}$ on average). DEHP was the dominant species, which constituted 42.2% of the total PAEs, followed by BBZP (146.8 ng $m^{-3}$ on average), which accounted for 15.4% of the total PAEs. DNOP, DEHA, and DBP were similar and each of them contributed for about 9% of total PAEs. DMP has the lowest concentration of 61.3 ng $m^{-3}$ on average, and constituted 6.4% of the total PAEs. DEHP has been found as the most abundant PAEs in ambient particle,

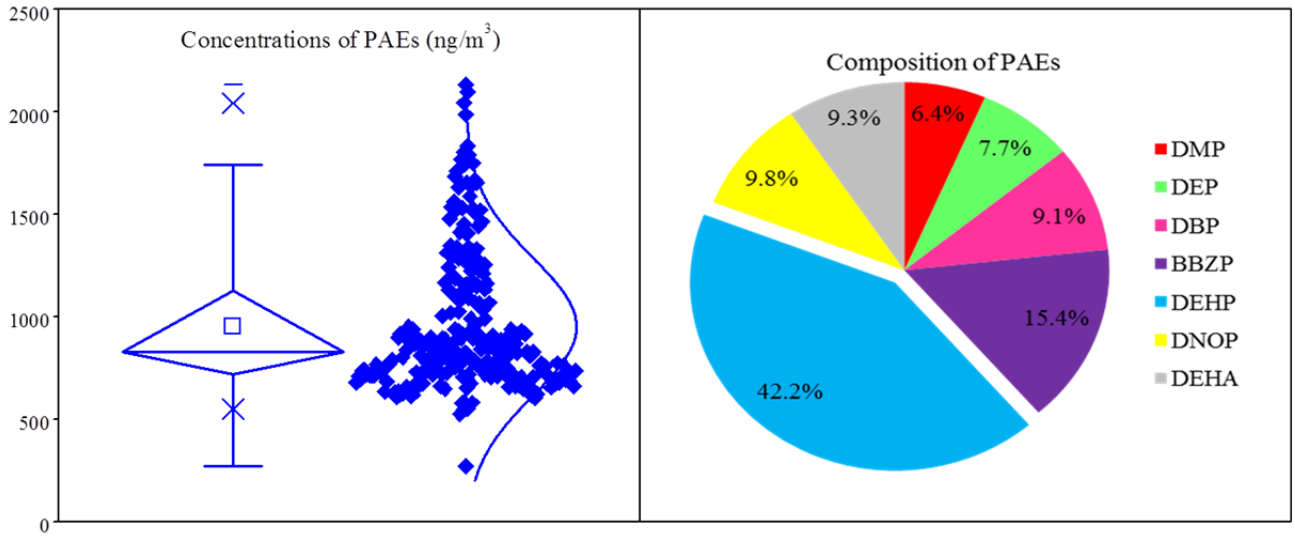


**Fig. 2.** The statistical concentrations of PAEs and percentages of different species at all of the sampling sites in $PM_{2.5}$ in Xi'an.

indoor dust and other matrixes for its wide usage in industry production (Zhu *et al.*, 2016). And it has been identified as the carcinogen so it is important to assess human exposure to PAEs especially to DEHP.

The worldwide comparison of PAEs in indoor air and outdoor particles were showed in Table 1. PAEs in China have much higher levels than in France, Korea, Greece and remote areas in Arctic and North sea (Teil *et al.*, 2006; Wang *et al.*, 2009; Xie *et al.*, 2005, 2007; Salapasidou *et al.*, 2011), but comparable with that in India (Fu *et al.*, 2010).

**Table 1.** Comparisons of PAEs in indoor and outdoor air at different regions and areas worldwide.

| Location | type | Sampling | Concentrations (ng $m^{-3}$) | Reference |
|---|---|---|---|---|
| Chongqing, China | Gas-phase, 30 residential | 2014 Nov.–2015 Feb. | 2380/1660[a] | Bu *et al.*, 2016 |
| Hangzhou, China | Gas-phase, 10 office | 2013 spring–fall | 2000–3800 | Song *et al.*, 2015 |
| Hangzhou, China | Indoor $PM_{2.5}$, 10 office | 2013 spring–fall | 774.6–2051 | Song *et al.*, 2015 |
| Tianjin, China | Indoor $PM_{2.5}$, 13 home | 2010 Dec; 2011 Jun | 7.2–1244 | Zhang *et al.*, 2014 |
| Xi'an, China | Gas-phase, 14 office and 14 home | 2012 Sep–2013 Jan | 200-8290 | Wang *et al.*, 2014 |
| Xi'an, China | TSP, 14 office and 14 home | 2012 Sep–2013 Jan | 90–14770 | Wang *et al.*, 2014 |
| Hangzhou, China | TSP, 10 new decorated residential | 2011 fall–2012 spring | 1067–14492 | Pei *et al.*, 2013 |
| Tianjin, China | $PM_{2.5}$, outdoor | 2010 spring, summer and winter | 85.6 | Kong *et al.*, 2013 |
| Tianjin, China | $PM_{10}$, outdoor | 2010 spring, summer and winter | 113.8 | Kong *et al.*, 2013 |
| Tianjin, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 127 ± 9.1 | Li *et al.*, 2015 |
| Tianjin, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 972 ± 132 | Li *et al.*, 2015 |
| Xi'an, China | TSP, outdoor | 2009 Jan–Feb | 268 | Li *et al.*, 2014 |
| Nanjing, China | TSP, outdoor | 2009 Apr–2010 Jan | 106 ± 8.2 | Wang *et al.*, 2012 |
| Hongkong, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 174 ± 33 | Li *et al.*, 2015 |
| Hongkong, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 407 ± 24 | Li *et al.*, 2015 |
| Guangzhou, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 335 ± 0.41 | Li *et al.*, 2015 |
| Guangzhou, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 440 ± 29 | Li *et al.*, 2015 |
| Xiamen, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 150 ± 29 | Li *et al.*, 2015 |
| Xiamen, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 205 ± 4.5 | Li *et al.*, 2015 |
| Wuhan, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 140 ± 28 | Li *et al.*, 2015 |
| Wuhan, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 477 ± 95 | Li *et al.*, 2015 |
| Chongqing, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 335 ± 91 | Li *et al.*, 2015 |
| Chongqing, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 1162 | Li *et al.*, 2015 |
| Shanghai, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 343 ± 200 | Li *et al.*, 2015 |
| Shanghai, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 623 ± 19 | Li *et al.*, 2015 |
| Xi'an, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 451 ± 14 | Li *et al.*, 2015 |
| Xi'an, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 673 ± 97 | Li *et al.*, 2015 |
| Qingdao, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 99 ± 20 | Li *et al.*, 2015 |
| Qingdao, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 170 ± 55 | Li *et al.*, 2015 |
| Beijing, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 179 ± 113 | Li *et al.*, 2015 |
| Beijing, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 408 ± 63 | Li *et al.*, 2015 |
| Changchun, China | $PM_{2.5}$, outdoor | 2003 Jan 13–14 | 117 ± 20 | Li *et al.*, 2015 |
| Changchun, China | $PM_{2.5}$, outdoor | 2003 Jun–Jul | 416 ± 97 | Li *et al.*, 2015 |
| Taizhou, China | $PM_{2.5}$, outdoor, 2 urban area | 2006 Jul | 106.2 (216.4) | Gu *et al.*, 2010 |
| Taizhou, China | $PM_{2.5}$, outdoor, 2 urban area | 2007 Jan | 197.1(326.6) | Gu *et al.*, 2010 |
| New Delhi, India | TSP, outdoor | 2006 Nov–2007 Feb, 2008 Jan | 884 | Li *et al.*, 2014 |
| Chennai, India | $PM_{10}$, campus | 2007 Jan–Feb | 175–598 (303) | Fu *et al.*, 2010 |
| Chennai, India | $PM_{10}$, campus | 2007 May | 295–857 (553) | Fu *et al.*, 2010 |
| Thessaloniki, Greece | $PM_{10}$, complex urban atmosphere | 2007 Jan–Feb | 23.0 (5.3)[b] | Salapasidou *et al.*, 2011 |
| Paris, France | TSP, outdoor, university | 2002 May–2003 Apr | 3.9–13.0 (8.2) | Teil *et al.*, 2006 |
| Gosan, Korea | TSP, outdoor | 2005 Mar–Apr | 8–47 (21) | Wang *et al.*, 2009 |
| Arctic | TSP, outdoor | 2004 summer | 0.38–1.02 (0.793) | Xie *et al.*, 2007 |
| North Sea | TSP, outdoor | 2004 Mar | 1.12–2.38 (1.66) | Xie *et al.*, 2005 |

[a] Living room and bed room, [b] traffic and industry.

The indoor air usually showed higher loaded concentrations than outdoor air, and indoor emission was a main source for outdoor PAEs (Kong *et al.*, 2013; Zhang *et al.*, 2014). The gas phase usually has higher levels than aerosol particles (Song *et al.*, 2015). However, it was regret that the gas phase was not collected in this study. Previous study has showed that large amount of PAEs existed in vapor form, especially for the low molecular weight and more volatile compounds with shorter dialkyl chains, such as DMP and DEP. DEHP and DNOP, the high molecular weight were high loaded in particles (Wang *et al.*, 2008). The partitions between the particle and gas phases of organic pollutants such as PAHs and PAEs also have been described somewhere (Wang *et al.*, 2008; Wei *et al.*, 2015). The particle-gas partition coefficient for individual PAEs associated with many factors, and played an important role for the fate of PAEs. We paid more attention on the health effect of these compounds, and the gas phase PAEs were not calculated here.

Li and Wang (2015) had detected $PM_{2.5}$ bound PAEs in 14 cities in China during the summer and winter at 2003. Higher PAE concentrations were obtained in summer unusually than in winter (Li and Wang, 2015; Zhu *et al.*, 2016). However in urban areas in Taizhou and in Tianjin, higher concentrations were found in winter (Gu *et al.*, 2010; Kong *et al.*, 2013). That is similar with the report by Wang *et al.* (2008), who found that PAEs decreased significantly with increasing temperature. This might due to the increase photochemical reactions in summer and enrichment of aerosol loaded in winter. Two of them, Xi'an and Chongqing, which has the basin-like topography, have much higher concentrations in wintertime. That was consistent with the heavy air pollution in winter for their low pollutants diffusion. Moreover, PAEs levels in Xi'an have been increased by 2 or 3 times in the past 10 years compared with Li and Wang (2015).

The impacts of meteorological conditions were also assessed here. PAEs have positive correlations with H, that the correlation coefficient ($R^2$) was 0.43. Ventilation coefficient (VC), which is calculated by the MLH multiply the U, has negative correlations ($R^2 = 0.58$) with pollutants (Kompalli *et al.*, 2014) (Supplemental Fig. S1). This was similar with the previous study about $PM_{2.5}$ bound PAHs in winter that H and VC are two main factors for the pollutants diffusion (Wang *et al.*, 2016a).

### *Spatial Distributions and Possible Sources of PAEs*

The spatial distributions of selected PAEs were presented in Fig. 3. Relatively high concentrations of the total PAEs were found in the southwest and in the city center. The southwest were the Xi'an high-tech industries Development Zone where the pharmaceutical companies, leather manufacturing, chemical plant, paper mills, household washing goods and cosmetic products plant, which might emit PAEs during the manufacture or add to the products. Intensive residential areas, commercial districts and heavy traffic might induce huge amount of PAEs in the city center. Lower PAEs levels were got in most of the north and northeast areas of the city, where there has the Xi'an International Horticultural Expo Garden and Ecological District, the Chan-Ba Ecological District, which provided a better air quality here (Xu *et al.*, 2016).

Fig. 4 illustrated that the PAEs concentrations declined along with the urban to suburban region. The average of total PAEs in the old cities wall, between the old city wall

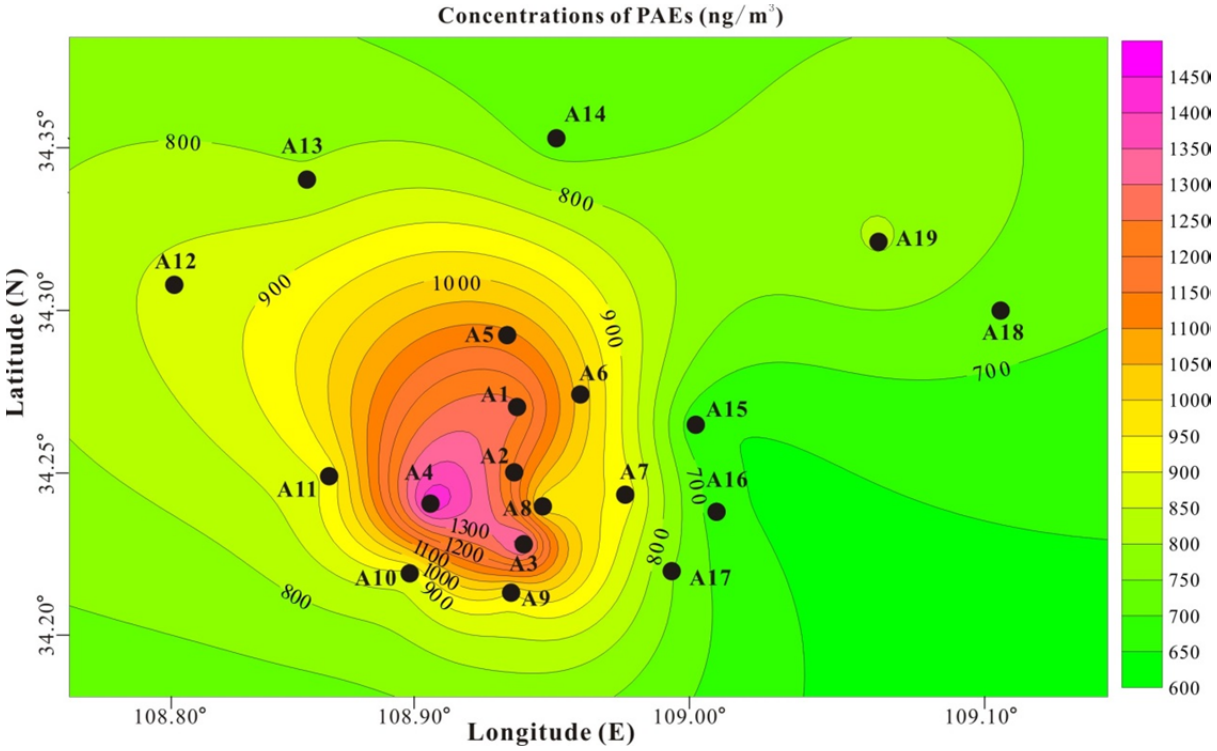


**Fig. 3.** Kriging interpolations of PAEs concentrations in Xi'an.

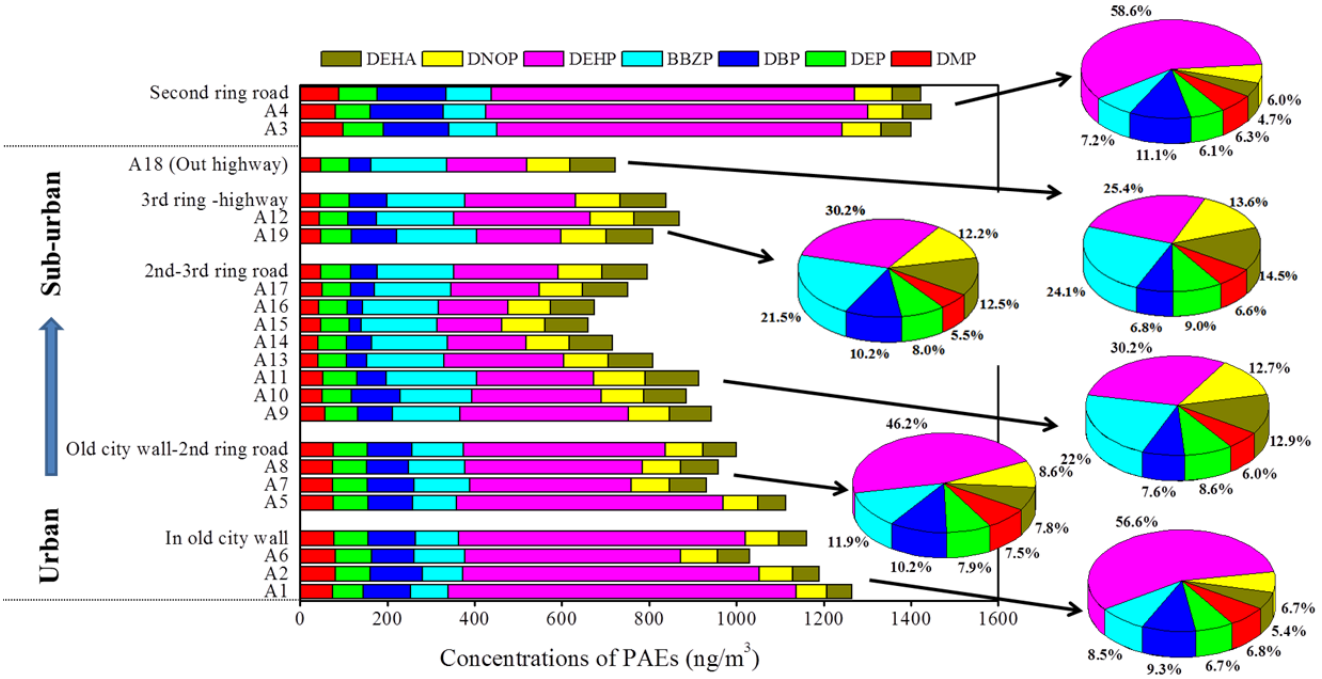


**Fig. 4.** The change of concentrations and compositions of PAEs from urban to suburban in Xi'an.

and 2nd ring road, between the 2nd and 3rd ring road, between 3rd ring road and highway, and out highway were 1161, 1000, 794.9, 838.8 and 722.0 ng $m^{-3}$. This demonstrated that the PAEs directly emitted from the products during its usage and disposal. The highest levels were got in site A3 and A4 (with an average of 1423 ng $m^{-3}$), which are near or very closely to the second ring roads, where have the heavy traffic pollution in Xi'an, showing that the tire wear might be a source for PAEs here. DEHP attributed for 56.6% of total PAEs in sampling sites in the old city wall, which decreased to 46.2% in sites between the old city wall and 2nd ring road, and 30.2% in the sites between 2nd and 3rd ring road. It has comparable contributions in the 3rd ring road-highway and 2nd- 3rd ring road. In the out highway (A18), DEHP was decreased to 25.4%. Correspondingly, BBZP was increased from 8.5% to 11.9%, then to 22.0% and 21.5% in the old city wall, the old city wall and -2nd ring road, and 2nd- 3rd ring road and 3rd ring-highway. It elevated to 24.1% in the out highway.

The coefficient of diversity (CD) was usually used for identifying the similarities of compounds between different sites (Wongphatarakul *et al.*, 1998; Xu *et al.*, 2016). The calculated formula and their meanings of the expressions can be obtained in the Supplemental materials. The CD value was between zero to one, and more approaches one meaning more significantly different between the two categories (Pongpiachan and Iijima, 2016). The CDs between different sites for PAEs in Xi'an were summarized in Supplemental Table S3. The CD values varied with the sampling sites, showing their different composition and sources. It ranged from 0.030 to 0.417 for all the sites. Previous study considered that when the CD values were < 0.269, they were predominantly affected by comparable sources (Wongphatarakul *et al.*, 1998). From Supplemental Table S3, it investigated that most of the CD values were lower than 0.269, which demonstrated that their similar sources. The CD values between A1–A5 and A11–A19, A6–A8 and A15, A16, were higher than 0.269, showing their difference of the sources. Though the compositions varied with sampling site (Fig. 5), DEHP was the dominant species, which accounted from 22.4% (A15) to 63.1% (A). Then followed by BBZP, which was from 6.8% (A1) to 26.4% (A15). They have the opposite trends, which DEHP was decreased whereas BBZP increased from the city central to the suburban. And the composition of PAEs was changed step by step from the city central to the suburban.

In this study, a principle component analysis (PCA) was performed using SPSS software to identify the possible sources for PAE (Table 2 and Supplemental Fig. S2). Two factors contributed for 86.8% of the total PAEs. Factor 1 consists of DNOP, DEHA, and BBZP, were attributed 43.8% of the variance, indicating the influence of plasticizers added in vinyl flooring, adhesives, synthetic leather, inks and food contact wrapping (Gómez-Hens and Aguilar-Caballos, 2003; Kong *et al.*, 2013; Silva *et al.*, 2013). Factor 2, including DMP, DEP, DBP, DEHP, accounted for 42.9% of the variance. That might be related with release from cosmetics and personal care products and volatilization from solid waste landfill or sewage and industrial wastewater. Previous study conducted by Cai *et al.* (2007) showed that DEHP contributed 94% of total PAEs in sewage sludge from Beishiqiao wastewater treatment plant in Xi'an, which located at the southwest of the city, and treat with mixture of domestic and industrial wastewater. It was comparable with the sewage sludge from two wastewater treatment plants in Hong Kong and Wuxi, but higher than in that in Beijing, Lanzhou, Guangzhou, and Shenzhen. Another research reported that DEHP accounted for 78% of sixteen

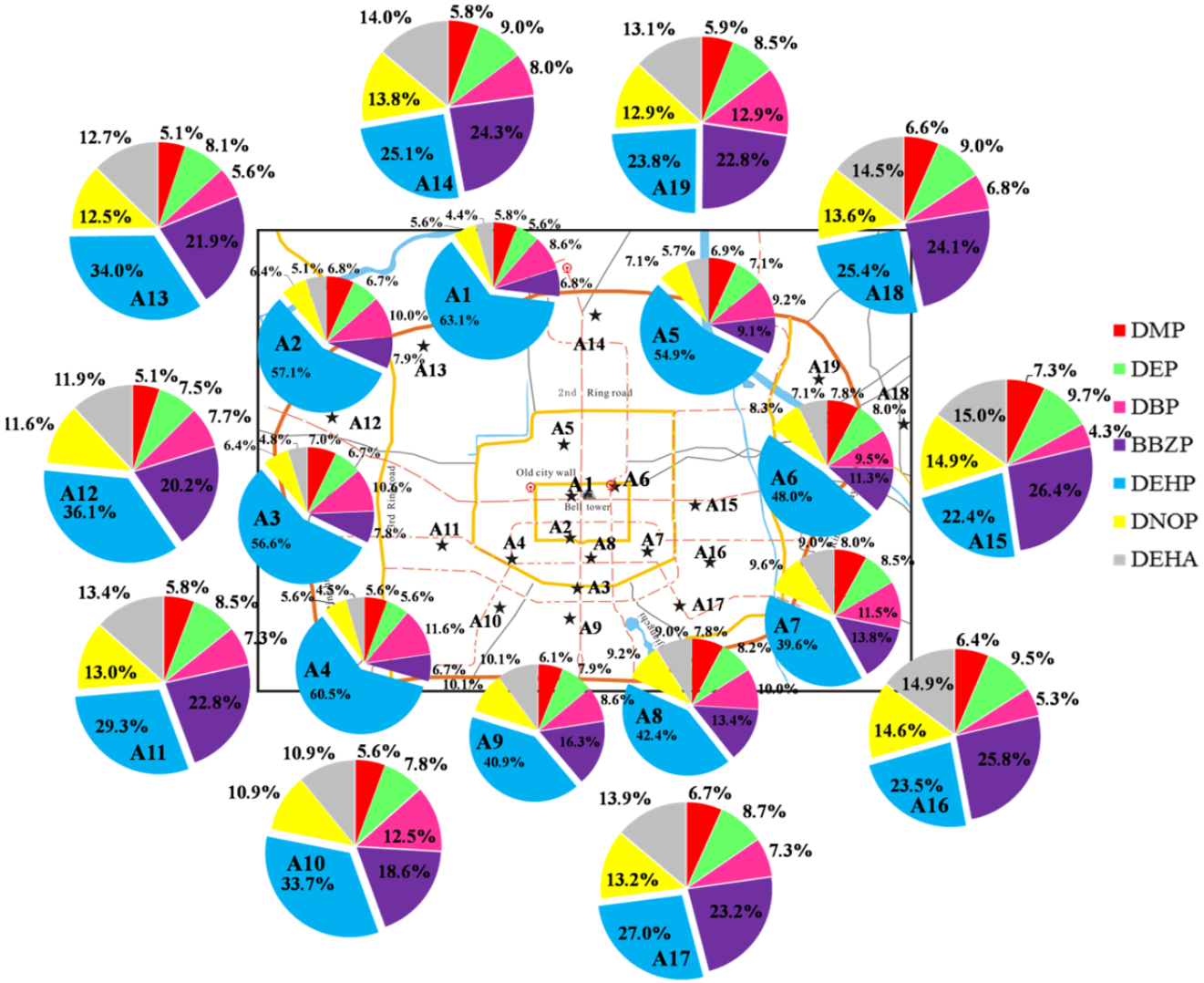


**Fig. 5.** Spatial distributions of PAE and compositions of PAEs at 19 communities in Xi'an.

**Table 2.** Principle component analysis for PAEs in $PM_{2.5}$ during haze periods at Xi'an.

| | PC1[a] | PC2 |
|---|---|---|
| DMP | –0.141 | 0.925 |
| DEP | 0.334 | 0.907 |
| DBP | –0.193 | 0.793 |
| BBZP | 0.950 | –0.292 |
| DEHP | –0.348 | 0.733 |
| DNOP | 0.984 | 0.155 |
| DEHA | 0.953 | –0.218 |
| Variance | 43.8 | 42.9 |
| Source | plasticizers added in vinyl flooring, adhesives, synthetic leather, inks and food contact wrapping | cosmetics and personal care products and volatilization from solid waste landfill or sewage from wastewater treatment plants |

[a] PC stands for principal component.

quantified PAEs in the surrounding environment of a municipal solid waste landfill in Wuhan, China (Liu *et al.*, 2010). For lack of specific emission sources for PAEs, it is difficult to effectively quantify the contributions of each source. So in order to better understand the sources of PAEs in the ambient air, the profile characteristic of PAEs emission sources should be investigated in the future.

***Health Risk Assessment***

Base on the inhalation exposure and carcinogenic risk

using DEHP, the estimated data for different age groups were summarized in Supplemental Table S4 and Fig. 6. It was clear that the average EI values for DEHP and BBZP were higher than the other individual PAEs for all age groups. This was associated with the concentrations of PAEs species at Xi'an. For the gas-phase PAEs were not monitored in this study, so the low molecular weight PAEs such as DMP, DEP, DBP, and BBZP might have been under-estimated. The total EI for PAEs in infants, toddlers, children, teenagers, and adults were 771.6, 342.9, 322.2, 226.5, 181.0 ng $kg^{-1}$ $day^{-1}$, respectively. And the decreased order for different age was: infants > toddlers > children > teenagers > adults. The EI for infants have the highest values, where adults have lowest estimated for all the PAEs species.

The estimated EIs for DMP and DBP via inhalation were lower than the indoor air estimated in Tianjin, Berlin and New York (Fromme *et al.*, 2004; Adibi *et al.*, 2008; Zhang *et al.*, 2014) for few researches were develop in ambient air. But the DEHP and DNOP were higher than the previous studies (Otake *et al.*, 2004; Wormuth *et al.*, 2006; Guo *et al.*, 2011b). The EIs of DEP, DBP, BBZP, and DEHP in this study were both lower than the reference values like the tolerable daily intake (TDI) by European Scientific Communities on Toxicity, Ecotoxicity and the Environment (CSTEE) (CSTEE (EU Scientific Committee on Toxicity) and US EPA's reference does (RfDs) (U.S. EPA) (Supplemental Table S4). This does not mean PAEs has low health risk, for much high exposure of DEHP in this area. The calculated cancer risk, basing on DEHP for age groups were showed in Supplemental Table S4. All of the average CR was higher than $10^{-6}$, which has potential health risks. The CR for the infants were from $1.09 \times 10^{-6}$ to $1.68 \times 10^{-5}$, showing that infants have higher health risks than the other age groups. For the non-estimated gas phase associated pollutants and other human exposure ways to PAEs, more researches need to be done for population in this area might cause concern for human health.

## CONCLUSIONS

The $PM_{2.5}$ bound PAEs were widely investigated in 19 communities in Xi'an, one of a heavy air pollution city in China. Much high levels of total PAEs were obtained, which from 271.7 to 2134 ng $m^{-3}$ (by an average of 952.6 ng $m^{-3}$). The spatial distribution showed that relatively high concentrations were found in the southwest and in the city center, and then decreased to the suburban. During all the sampling sites, DEHP was the dominant species followed by BBZP. DEHP was decreased whereas BBZP increased from the city central to the suburban. Principle component

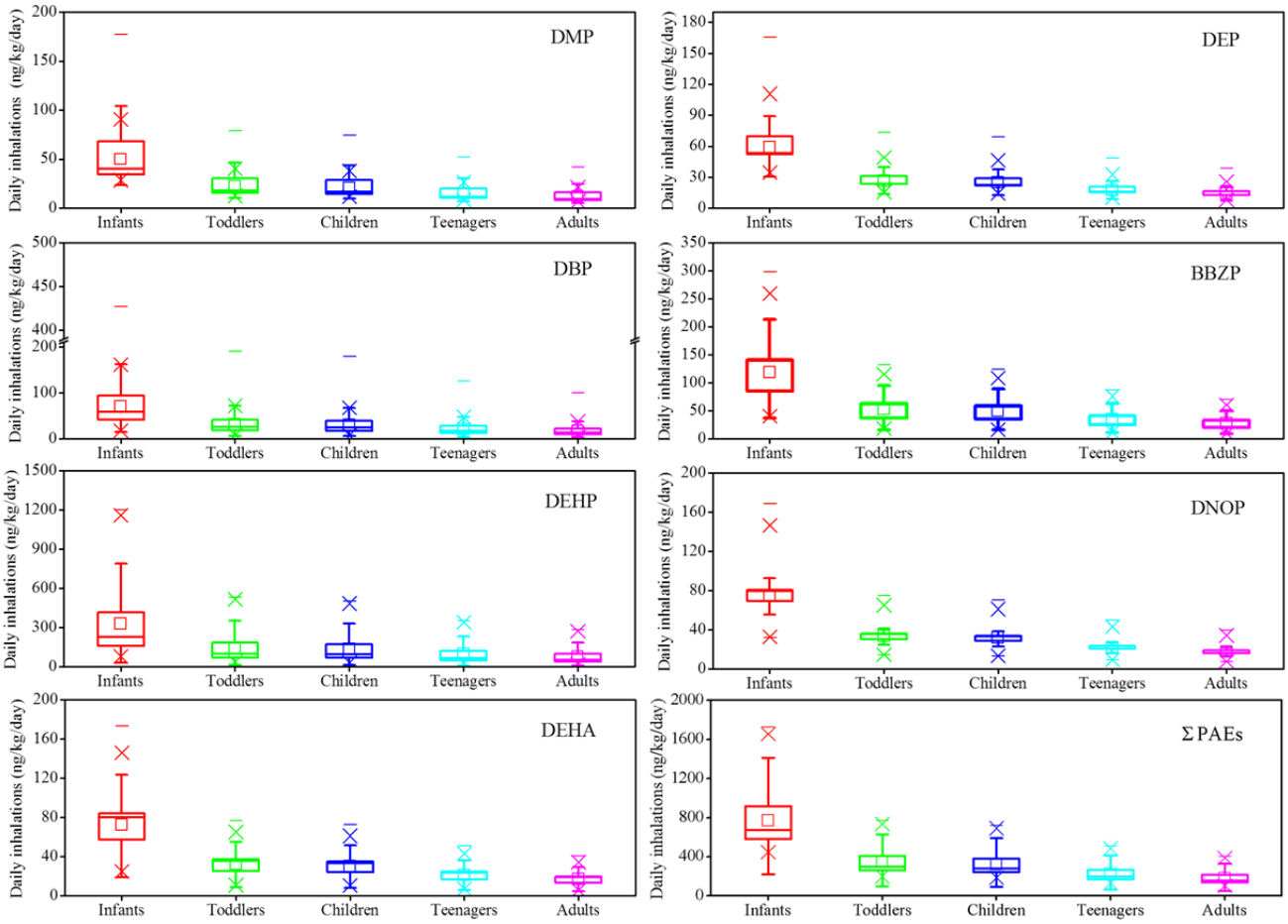


**Fig. 6.** Estimated daily inhalations of PAEs (ng $kg^{-1}$ $day^{-1}$) for five age groups in Xi'an. Box-whisker plots respects the 5th, 25th, 50th, 75th, and 95th percentiles, as well as the mean (open squares).

analysis demonstrated that plasticizers added in vinyl flooring, adhesives, synthetic leather, inks and food contact wrapping (by usage), and the volatilization from waste stacks or sewage and industrial wastewater (by disposal) were two main sources for PAEs. The cancer risk assessment investigated infants as the susceptible people have more potential risks than the other age groups. So it is important to fully understand the exposure to PAEs in this area.

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## SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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