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ARTICLE

The cumulative characteristics of PAEs in $PM_{2.5}$ in Changji, Northwest China

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ABSTRACT

This paper discusses characteristics of the seasonal variation in phthalate acid esters (PAEs) in the $PM_{2.5}$ in the city of Changji, which is located an arid area of Xinjiang Province, northwest China. Samples were collected using a Laoying 2030 median discharge intelligent total suspended particulate (TSP) sampler and analyzed by a Shimadzu GC-2010 gas chromatograph. The results showed that PAEs in $PM_{2.5}$ were mainly composed of phthalic acid dibutyl phthalate (DBP) and phthalic acid (2-)ethyl hexyl ester (DEHP). DBP and DEHP are the most commonly used plasticizers, accounting for the majority of plasticizers used, and have large environmental emissions. Compared with the proportions of TSPs, proportions of DBP and DEHP in the $PM_{2.5}$ were lower, while the proportions of DMP and DEP were higher. A correlation exists between the distribution of PAEs and the particle size and concentration of atmospheric particulates. Seasonal variations in DBP and DEHP were the same, with lowest concentrations in the summer. Although seasonal variation in DMP was not the same as those of DBP and DEHP, the general trend was similar. Seasonal variation in DEP was different from those of other PAEs. This pattern is related to PAE characteristics and seasonal variations in their sources.

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Arid areas; $PM_{2.5}$; PAEs; Accumulation

Introduction

Air pollution in cities is an extremely serious problem in contemporary China, and public attention to air pollution is increasing. Different from coarse particulate matter, $PM_{2.5}$ is not easily removed through dry deposition because of its small size, low weight, and large quantity. Thus, it can stay in the atmosphere for a long time and travel long distances through atmospheric circulation, leading to regional air pollution (Hassana & Khoderba, 2017; Ryswyk et al., 2017). Furthermore, its smaller particles can more easily enter the human respiratory system. Atmospheric particulate matter with diameters <2.5 μm ($PM_{2.5}$) can directly enter the lungs, and nearly 75% of $PM_{2.5}$ is deposited in the alveoli, causing serious damage (De, Groth, & Veras et al., 2018; Nemmar et al., 2002). A large number of epidemiological and toxicological studies have confirmed that $PM_{2.5}$ pollution is significantly associated

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with mortality, systematic respiratory issues and cardiovascular morbidity (Dunea et al., 2016; Ma et al., 2014; Polezer et al., 2018). Pope, Burnett, and Thun et al. (2002) found that the numbers of patients with heart and lung disease and lung cancer increase by 6% and 8% for every 10 $\mu g/m^3$ increase in $PM_{2.5}$ concentration, respectively. Compared with PM_{10}, $PM_{2.5}$ poses a greater threat to human health (Li, Zhang, Qiu, Wang, & Fang, 2017).

Phthalate acid ester (PAE) compounds have the characteristics of teratogenicity, carcinogenicity, mutagenicity, and especially reproductive toxicity. The persistent harm caused by these compounds is severe (Bui et al., 2016; Sakhi et al., 2014; Xia, Ouyang, Wang, Shen, & Zhan, 2018). PAEs are mainly used as plasticizers in the production of plastic products to enhance their plasticity and strength. PAEs are connected to plastic molecules by hydrogen bonds or van der Waals forces with relatively poor stability. Thus, during the production, use, and final disposal of plastic products, PAEs readily escape from the plastic into the environment (Al-Natsheh, Alawi, Fayyad, & Tarawneh, 2015; Kong, Kadokami, Wang, Duong, & Chau, 2015). PAEs have been widely detected in natural and human environments in China, such as the atmosphere, water, soil, organisms, and the human body (Liu, Tian, Liu, Liu, & Liu, 2016a; Palusel et al., 2017; Alsaleh, Elkhatib, Alrajoudi, & Al-Qudaihi, 2017). In the 1980s, Zhao and Xu (1982) found that the concentrations of phthalic acid dibutyl phthalate (DBP) and phthalic acid (2-)ethyl hexyl ester (DEHP) in atmospheric particulates in some areas of Beijing were 10 to 50 times higher than those in uninhabited areas. At the beginning of the twenty-first century, Wang, Wang, & Fan (2008) found that the concentrations of PAEs in the center of Nanjing were 3.5 times higher than those in the suburbs. In recent years, Liu et al. (2016a) found that the concentrations of dimethyl phthalate (DMP), diethyl phthalate (DEP), DBP, and DEHP in urban soil samples of Changji (Xinjiang) were significantly greater than the United States soil PAE compound control standards to different degrees; in particular, the DBP concentration exceeded the standard by >3 orders of magnitude (Wang et al., 2008). The US Environmental Protection Agency (EPA) assigned the following six PAE compounds to its key control pollutant list: DMP, DEP, DBP, butyl benzyl phthalate (BBP), DEHP, and dioctyl phthalate (DOP). Moreover, DMP, DBP and DOP are also included on the list of environmental priority pollutants in China.

The toxicity of particles is closely related to the particle size. In addition to the fact that smaller particles can more easily enter the human respiratory system, they have higher specific surface areas, making it easier for various pollutants to be absorbed in the air (Liu et al., 2016c). The synergistic effect of atmospheric particulates and pollutants can produce stronger toxicity (Mesquita et al., 2016; Singh et al., 2010). The scientific community has carried out relevant studies on PAEs in atmospheric particulates and accumulated a significant amount of data. Some reports are also available on the seasonal variation characteristics of PAEs in $PM_{2.5}$ (Gupta & Gadi, 2018; Lu et al., 2018). However, PAE accumulation, seasonal variation, and association with $PM_{2.5}$ in arid areas remain unclear. Meteorological and climatic conditions are the main factors affecting the migration and transformation of urban environmental pollutants (Liu et al., 2016c). The meteorological and climatic conditions in arid areas are special, and the effects of these conditions on the accumulation of PAEs in $PM_{2.5}$ have not been studied in depth. Thus, investigating the accumulation characteristics of PAEs in the urban $PM_{2.5}$ in Changji, an arid area of Xinjiang, is of importance to understanding the theory and regional characteristics of PAEs in arid environments.

Materials and methods

Sample collection

Changji (44.05°N, 87.31°E) is located in an arid to semiarid region on the Eurasian continent. It is a satellite city of Urumqi, the capital city of Xinjiang. It has a typical continental dry climate with an annual precipitation of only ~200 mm. Changji has four distinct seasons, with a winter that is cold and long, requiring a 6-month heating period from October to April. With its fast economic development, Changji has experienced rapid urbanization in recent years. The increase in atmospheric pollutant emissions is mainly due to coal burning during the winter for heating and spring dust storms. The average annual concentrations of $PM_{2.5}$ in the years from 2015 to 2018 were 40.5 μg/m^3, 42.4 μg/m^3, 49.9 μg/m^3, and 50.2 μg/m^3, respectively. Air quality is declining, and the urban ecological environment is facing unprecedented pressure.

Three sampling sites were established based on urban functional zoning and sampling microenvironments in Changji: an old school district of Changji University (residential and educational), the Changji passenger station (traffic area), and the Lanshandtunhe Company (industrial area). The sample sites are not surrounded by local pollution sources or tall trees or buildings. The sampling height was 15 m above the ground. Samples were collected for 3 days in the middle of each month from April 2017 to March 2018. If the scheduled sampling campaign coincided with rainy or windy weather (wind over 4), the sampling was postponed for 3 days after the precipitation and wind. $PM_{2.5}$ and TSP (total suspended particulate) samples were collected synchronously and continuously for 6 h using two Laoying 2030 Medium Flow Rate Intelligent TSP Samplers (Laoying Institute, Qingdao, SD, China) at a flow rate of 100 L/min. One sampler collected $PM_{2.5}$, while the other collected TSP. Temperature, air pressure, wind speed, wind direction, humidity, and sample quality were recorded synchronously. TSP data were collected at the same time for the preliminary discussion of the PAE particle size effect. No component had any contact with plastic products throughout the sampling process.

Sample analysis

Main instruments and reagents

The instruments included a Laoying 2030 Medium Flow Rate Intelligent TSP sampler (Laoying Institute, Qingdao, SD, China) (Liu et al., 2018), a Shimadzu GC-2010 gas chromatograph, an RE-52AA rotatory evaporator and an HY-CXJ high-power digital ultrasonic cleaner. The reagents included PAE standard samples of DMP, DEP, DBP, and DEHP with concentrations of 1.0 mg/mL, n-hexane, and acetone.

Analysis method

PAEs were extracted using a Soxhlet extractor for 10 h. N-hexane/acetone (1:1) was used as the extracting agent. We used gas chromatography-flame ionization detection (GC-FID) to determine the PAE concentrations. Among the main working parameters of GC-FID, the injector and ion source temperatures were maintained at 260°C. The nitrogen carrier gas (≥99.999%) was kept at a constant flow of 30 mL/min, the hydrogen gas (≥99.999%) was kept at a constant flow of 40 mL/min, and the air (≥99.999%) was

kept at a constant flow of 400 mL/min. The ratio of hydrogen to airflow is approximately 10:1. The extracted samples were injected into the gas chromatography (GC) in split mode with a split ratio of 15:1.

Quality assurance and quality control

Measurements of mixed standard solutions of 50 μg/mL were repeated five times to test the precision of the methods. The results showed that the relative standard deviations of the method were all less than 5%, at 2.0%, 1.6%, 2.2%, and 4.2% for DMP, DEP, DBP, and DEHP, respectively. To ensure the reliability of the data, the same samples were measured in parallel, and the deviations were not greater than 5%.

Results and discussion

Concentrations of PAEs in $PM_{2.5}$

The average concentrations of DMP, DEP, DBP and DEHP in $PM_{2.5}$ were 6.31 (not detected (ND)–19.53) ng/m^3, 13.51 (ND–57.99) ng/m^3, 49.05 (2.75–175.57) ng/m^3, and 45.55 (3.24–259.86) ng/m^3, respectively. DBP and DEHP had the highest concentrations among the four PAEs, followed by DEP and DMP. DBP and DEHP account for 83% of the four PAEs, indicating that they are the main PAEs in the $PM_{2.5}$ in Changji. Some studies suggested that the main PAEs present in urban soil, sediment, and other media and on impervious surfaces are DBP and DEHP, and that there are similarities in the distributions of the PAEs in arid areas (Figure 1) (Liu et al., 2018, 2016a). DBP and DEHP are the most commonly used plasticizers, accounting for the majority of the plasticizers used, with large environmental emissions (Edjere, Asibor, & Otolo, 2016; Wang et al., 2017). In addition, due to the semivolatile characteristics of PAEs, they exist in both the gas phase and granular phase in the air. PAE adsorption usually increases with molecular weight. Therefore, relatively high molecular weight PAEs (such as DEHP and DBP) in the atmosphere are often attached to particles, while other PAEs with lower molecular weights exist in the vapor phase (Howard, Baberjee, & Robillard, 1985). These factors may be why DBP and DEHP are the major PAEs in $PM_{2.5}$.

Comparison of the structures of the paes in TSP and $PM_{2.5}$

The sum of DMP, DEP, DBP, and DEHP (Σ_4PAE) accounts for 7.44%, 16.84%, 43.43%, and 32.29% of the total PAEs in $PM_{2.5}$, respectively. PAEs in the TSP were analyzed to explore the particle size effect of the PAEs in the atmospheric particulates. The concentrations of DMP, DEP, DBP and DEHP were 2.50 ng/m^3, 2.59 ng/m^3, 82.58 ng/m^3, and 71.56 ng/m^3, respectively, accounting for 1.57%, 1.57%, 51.89%, and 44.97% of the total PAEs, respectively (Figure 2). Although PAEs in both the $PM_{2.5}$ and TSP are dominated by DBP and DEHP, their proportions were larger in the TSP; in contrast, the proportions of DMP and DEP were higher in the $PM_{2.5}$ than in the TSP.

A correlation exists between the distribution of the PAEs and the particle size and concentration of the atmospheric particulates (Teil, Blanchard, & Chevreuil, 2006). The particle size of TSP is ≤100 μm, while that of the fine particles is ≤2.5 μm. Generally, PAEs with higher molecular weights have a stronger affinity for adsorption and are

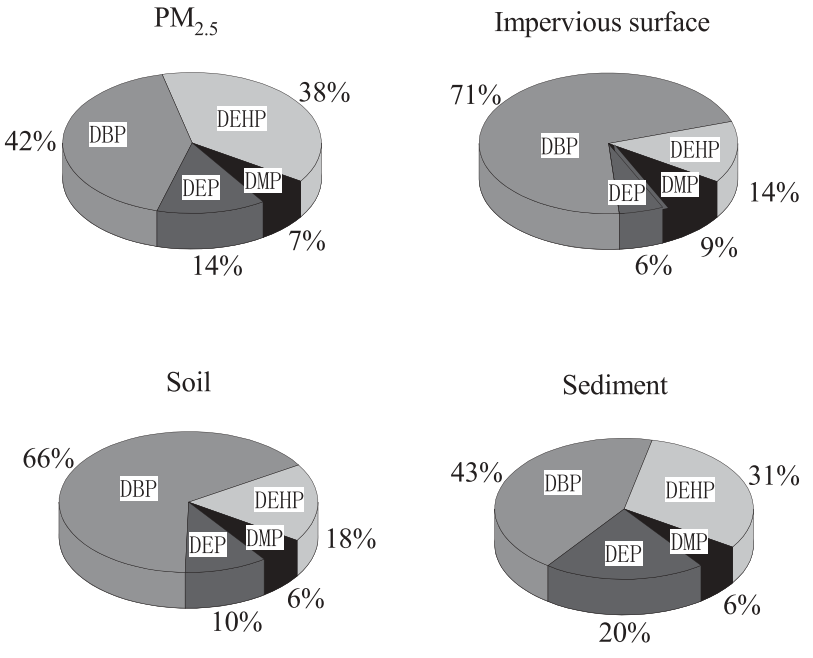

Figure 1. Compositional characteristics of $PM_{2.5}$ and of PAEs in urban soil, in sediment, and on impervious surfaces in an arid area.

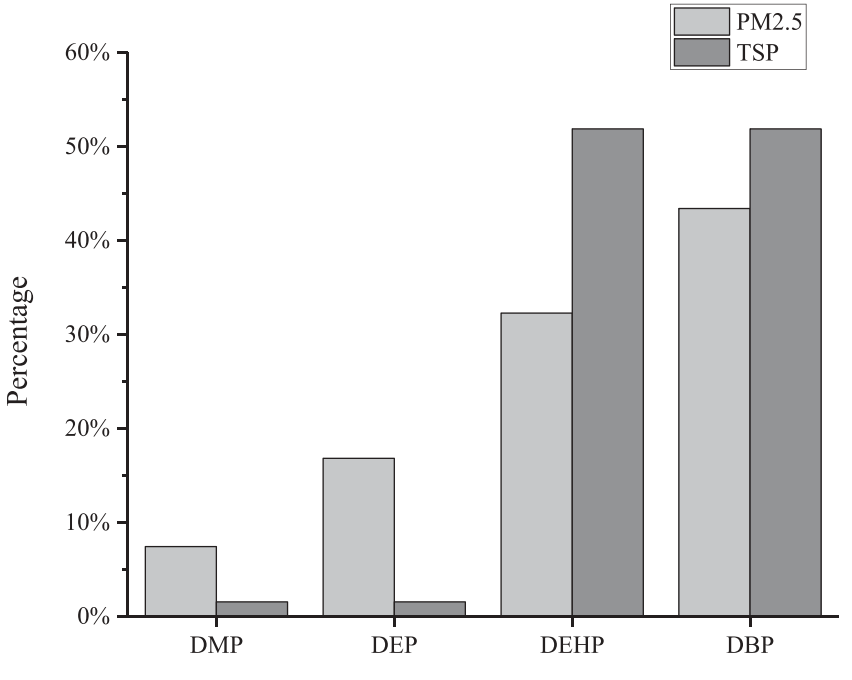

Figure 2. Compositional characteristics of DMP, DEP, DEHP, and DBP in $PM_{2.5}$ and TSP (total suspended particulates).

more likely to be adsorbed on the surface of particles (Jing, Pan, & Chen, 2018). Moreover, the highly adsorbent macromolecular PAEs will be adsorbed onto coarse particles with large surface areas, whereas the smaller PAE molecules are easily accumulated on fine particles, except in the presence of steam (Ma et al., 2014). Ma et. al. (2014) found that DEHP is the main PAE on large particles, while the proportions of DMP and DEP are high for fine particles, in agreement with the results of this study.

Seasonal variation

DMP concentrations in the $PM_{2.5}$ in the spring, summer, autumn, and winter were 6.44 ng/m^3, 5.93 ng/m^3, 4.75 ng/m^3 and 8.36 ng/m^3, respectively, and the DEP concentrations were 8.70 ng/m^3, 16.34 ng/m^3, 10.34 ng/m^3, and 14.51 ng/m^3. DBP concentrations were 94.59 ng/m^3, 23.66 ng/m^3, 45.61 ng/m^3, and 49.27 ng/m^3, respectively, and DEHP concentrations were 112.66 ng/m^3, 18.80 ng/m^3, 27.07 ng/m^3, and 41.48 ng/m^3. Seasonal variations in DBP and DEHP were similar, with the lowest concentrations in the summer and the highest in the spring. Although the seasonal variation of DMP was different from those of DBP and DEHP, the general trend was similar, with higher concentrations in spring and winter and lower concentrations in summer and autumn. Seasonal variation of DEP was different from those of the other PAEs: the concentration was highest in summer and lower in the other seasons (Figure 3).

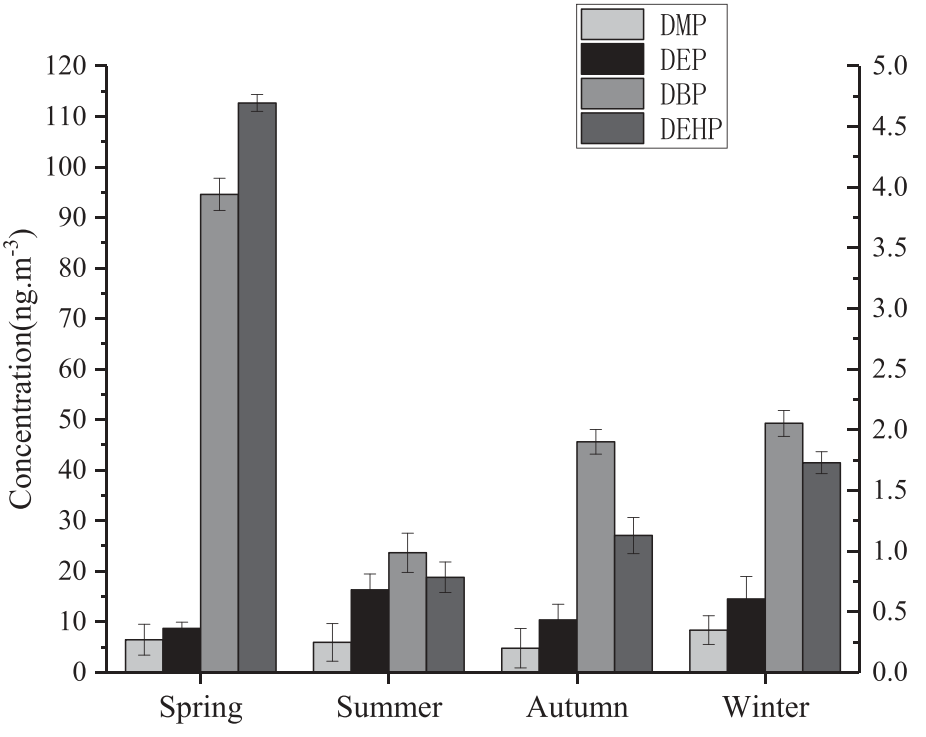

Figure 3. Seasonal variations in DMP, DEP, DBP, and DEHP in $PM_{2.5}$.

Similar to other studies, this study found two opposite patterns in the seasonal variations of PAEs. Focusing on PM_{10} and $PM_{2.5}$, Ma (2014) showed that PAE measurements in warm months were significantly higher than those in cold months and that PAE concentrations in summer were much higher than those in winter. PAEs are semivolatile pollutants; thus, an increase in the ambient temperature will directly affect PAE emissions from various plastic products, leading to higher concentrations of PAEs in the atmosphere in warm months (Clausen, Liu, Kofoed-Sørensen, Little, & Wolkoff, 2012). However, Wang et al. (2008) showed that concentrations of PAEs in the environment decrease with the increase in temperature and that PAE concentrations in summer are lower than those in winter. This finding is observed because photochemical reactions occur in the summer, thereby consuming PAEs and reducing their concentrations (Gupta & Gadi, 2018; Wowkonowicz & Kijeńska, 2017). The relationship between the degradation ability of PAEs and their structures has been discussed previously: Meylan and Howard (1993) believed that longer alkyl chains in PAE compounds are more susceptible to photodegradation reactions, and Ma et al. (2014) found that concentrations of DEHP, which has long alkyl chains, were negatively correlated with temperature in $PM_{2.5}$ and PM_{10}.

DBP and DEHP concentrations in the $PM_{2.5}$ in Changji City were lowest in the summer and the highest in the spring. The long alkyl chains of DBP and DEHP in the atmospheric particulates are prone to photolysis and loss in the summer, resulting in the lowest concentrations being in summer. Changji City has a long heating period covering the winter, spring, and autumn, and coal-fired heating increases the emissions of PAEs in these seasons (Chen, Lv, Li, & Zhu, 2018). In addition, spring is the main season of dust storms in Changji City. Dust storms pass through the main industrial areas of Xinjiang (Zhundong Development Zone) and carry the most commonly used plasticizers DBP and DEHP, which are released as industrial emissions into Changji. The above reasons led to relatively high DBP and DEHP concentrations in the winter, spring, and autumn, with the highest values in the spring. Note that high temperatures in summer can also increase the release of PAEs from plastic products. DMP concentrations in the $PM_{2.5}$ in Changji City were higher in spring and winter and lower in summer and autumn. The temperature of Changji in summer and autumn is higher than that in spring and winter. Concentrations of PAEs in the environment decrease with an increase of temperature (Wang et al., 2008), which results in the concentrations of DMP in summer and autumn being lower than in spring and winter. Higher concentrations of DMP in spring and winter may be related to PAEs emissions from coal-fired heating (Sun et al., 2014). However, compared with medium- and long-chain DBP and DEHP, DMP and DEP, with relatively low molecular weights and short alkyl chains, are more active (Langer et al., 2010; Li et al., 2016), more sensitive to high temperatures, and more easily separated from plastic products or volatilized from other media into the atmosphere. The behavior of DEP is particularly distinct from that of the others. This pattern is also the main reason that the DEP seasonal variation in $PM_{2.5}$ in Changji City is different from that of the other PAEs, with high concentrations in the summer and lower concentrations in the other seasons.

In summary, according to the length of the alkyl chain, PAEs can be divided into two types: medium- and long-chain PAEs (such as DBP and DEHP) and short-chain PAEs (such as DEP). Concentrations of medium- and long-chain PAEs in the $PM_{2.5}$ decrease

in summer due to the influence of summer photolysis and other factors. Short-chain PAEs are sensitive to high summer temperatures, and their emissions from plastics and other products increase. Finally, concentrations of short-chain PAEs in $PM_{2.5}$ increase in the summer. The differing conclusions of previous studies regarding seasonal variations in the PAEs were actually the result of examining different PAEs. When the same PAEs are studied, the results are consistent. Based on a comparison of seasonal variation characteristics of PAEs in other media in arid areas, it has been found that the seasonal variation in DEP in urban soil and on impermeable surfaces was significantly different from those of the other PAEs (Liu et al., 2016a), which may be related to the PAE characteristics and seasonal variations in their sources.

Conclusion

In summary, DBP and DEHP accounted for 83% of the four PAEs, indicating that they were the main PAEs in the $PM_{2.5}$ in Changji. Some studies have suggested that the PAEs present in the soil, sediment, and other media and on impervious surfaces are mainly DBP and DEHP and that there are similarities between the distributions of the PAEs in urban arid areas. DBP and DEHP are the most commonly used plasticizers, accounting for the majority of the plasticizers used, and they have large environmental emissions.

PAEs in both the $PM_{2.5}$ and TSP were dominated by DBP and DEHP. The proportions of DBP and DEHP are higher in TSP and lower in $PM_{2.5}$, while the proportions of DMP and DEP were higher in $PM_{2.5}$ than in TSP. DBP and DEHP, with relatively high molecular weights, preferentially adsorbed to coarse particles with large surface areas, while DMP and DEP, with relatively low molecular weights, more easily adsorbed onto fine particles. There is a certain correlation between the distribution of PAEs and the particle size and concentration of atmospheric particulates.

Seasonal variations in DBP and DEHP concentrations were similar, with the lowest values in summer and the highest in spring. Although the seasonal variation in DMP was different from those of DBP and DEHP, the general trend was similar, with higher concentrations in spring and winter and lower concentrations in summer and autumn. Seasonal variations in DEP differed from those of the other PAEs, with the highest concentrations in summer and lower concentrations in the other seasons. This pattern is related to PAE characteristics and seasonal variations in their sources.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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