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How human activities in commercial areas contribute to phthalate ester pollution in street dust of Taiwan

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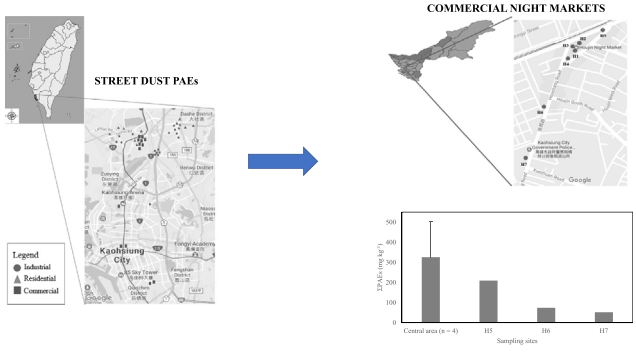
HIGHLIGHTS

- PAE levels in street dust are related to commercial activities.
- PAEs were detected in all samples in the range of 5.4–989.2 mg·kg^{-1}.
- Estimated PAEs non-dietary DIs are higher in children than those in adults.
- Night markets contributed considerably to street dust PAE contamination.

GRAPHICAL ABSTRACT



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ABSTRACT

Exposure to phthalate esters (PAEs) poses health risks to humans. Much research has been performed evaluating PAE levels in foodstuffs, river sediment and drinking water, but little attention has been paid to their presence in urban outdoor environments where human activities are highly intense. Here we evaluated PAE presence and distribution in street dust in Kaohsiung, the most industrialized city in Taiwan. Our results showed that PAEs were ubiquitous in fifty-two street-dust samples (levels of total PAEs 5.4–989.2 mg kg^{-1}). Di-(2-ethylhexyl) phthalate was the most abundant congener observed and made up 85.0%, 79.7%, and 97.2% of the total PAEs found in industrial, residential and commercial areas, respectively. PAE levels in street dust in commercial areas (night markets) were significantly higher, suggesting a higher risk of contamination on people present in these areas (H value > χ^2_U). In residential and commercial areas, the higher the intensity of human activity, the higher the PAE content observed. PAE content decreased progressively from the center to the outskirts of the Houjing night market, suggesting that the increased human and consumer activities inside this commercial hotspot were the main PAE source in street dust. Children had higher estimated daily intakes (DIs) than adults and dermal absorption contributed more to these levels than oral ingestion. Although all calculated DIs were below referenced danger thresholds, street dust PAEs in the area should remain an environmental concern especially since night markets play an important role in Taiwanese/Asian culture and economy. Contrary to other studies, PAEs in this study were found less related to industrial manufacturing activities but highly linked to commercial activities. These findings are relevant for future pollution prevention efforts dedicated to mitigating public exposure to PAEs.

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Main findings: PAE levels in street dust are related to commercial activities. Night markets, an important commercial activity in Taiwan, were found to contribute considerably to PAE contamination in street dust.

1. Introduction

Phthalate esters (PAEs), flexible and elastic chemicals, are used widely as plasticizers, particularly in products made of polyvinyl chloride (PVC) (Niu et al., 2014). They are found in a wide variety of daily products including children's toys, pesticides, food packaging, household furnishings, personal care products, medicines and even foodstuffs (Niu et al., 2014; Skrbic et al., 2016). One hundred fifty million tons of plastic is manufactured globally every year and PAEs are consumed at a rate of 6.0 to 8.0 million tons annually (Niu et al., 2014). As a result, PAEs are widespread in soil, water, sediments and air (Zeng et al., 2008b).

PAEs are classified as endocrine disruptors that pose health risks to both terrestrial and aquatic species (Zeng et al., 2008a; Niu et al., 2014). Exposure to these substances leads to various adverse health complications (Kozumbo et al., 1982; Kluwe, 1986). Six specific PAEs (dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (D*n*BP), benzyl butyl phthalate (BBP), di(2-ethylhexyl) phthalate (DEHP), di-n-octyl phthalate (D*n*OP) are highly toxic and are categorized as priority contaminants by the US Environmental Protection Agency (USEPA, 2013) and the European Union (EU) (Zeng et al., 2008b; Wu et al., 2012). Therefore, PAE contamination and propagation are often the subject of worldwide monitoring studies (Niu et al., 2014; Skrbic et al., 2016).

Kaohsiung City is the most industrialized area in Taiwan. It has a population of around three million and is an important economic and industrial center in Asia. In Kaohsiung, PAEs have been found in river sediment (Lin et al., 2009), drinking water (Yang et al., 2014), soil (Kaewlaoyoong et al., 2018) and foodstuff (Wu et al., 2013). PAEs were also detected in human urine in studies performed following a scandal involving industries that used PAEs as a substitute of emulsifier to food products they manufactured (Chen et al., 2016). Since that time, PAE contamination and associated health risk have been a major concern of Kaohsiung City government and its citizens (Yang et al., 2014).

A large number of studies have investigated PAEs in indoor dust (Bornehag et al., 2005; Hwang et al., 2008; Orecchio et al., 2013; Zhang et al., 2013; Ait Bamai et al., 2014; Jeon et al., 2016; Albar et al., 2017; Larsson et al., 2017; Subedi et al., 2017), and human incidences of PAEs from indoor contamination have been reported (Pei et al., 2013; Tran et al., 2016; Albar et al., 2017). However, while indoor contamination can be controlled or managed by inhabitants, PAE street dust exposure is less easily controlled (Lan et al., 2012; Skrbic et al., 2016). Although street dust PAE levels can serve as an important indicator of contamination, their presence in outdoor matrices is not usually monitored and is, therefore, not well documented (Zhang et al., 2013; Skrbic et al., 2016). To date, only a few studies of street dust PAE contamination have been reported, in China and Serbia (Lan et al., 2012; Zhang et al., 2013; Skrbic et al., 2016) and no studies have measured PAE contamination in street dust of Taiwan. Here, the industrialization has led to the rapid development of areas where plastic and metal processing industries thrive for the last forty years. The most industrialized city in Taiwan is Kaohsiung City, home to giant metal-plating and plastic industrial plants (Vu et al., 2017a; Vu et al., 2017b; Lee et al., 2018).

Like many other Asian countries, Taiwan has many night markets. There are around three hundred night markets operating in multiple cities throughout Taiwan and they are considered a signature of the Taiwanese/Asian culture (Sun et al., 2012; Chen et al., 2014). Night markets exist in almost every area of Kaohsiung City, and there are thousands of people working in or visiting these night markets every day (Zhao et al., 2011; Chen et al., 2014; Liao, 2017). Levels of PAEs in street dust in these night market areas, therefore, deserve to be studied because they could pose serious concerns on human health and wellbeing.

As mentioned above, investigations in outdoor dust are very scarce, as well as studies showing any link between these PAE contamination occurrences and the intensity of outdoor human activities. Therefore, this study aims to analyze the occurrence of PAEs in samples collected in Kaohsiung city to determine zones of high PAE contaminations in street dust. Daily intakes from the level of exposure to PAEs in street dust in both adults and children through ingestion and dermal pathways were estimated and pollution prevention strategies suggested.

2. Materials and methods

2.1. Study location and sample collection sites

Kaohsiung city, situated in southern Taiwan, is one of the most rapidly growing industrial cities in Asia (Chia-Hong, 2013). We collected a total of 52 street dust samples from three industrial, three residential and five commercial areas in the city from November 2014 to May 2015. The samples were collected on sunny days between 9 am and 4 pm. There was no precipitation at any of the sites two weeks before the sampling. Most street dust samples were taken within one week. Daily variations of street dust PAEs during a week of sampling was considered outside the scope of this study and were assumed to be negligible. Sampling sites selected were areas potentially contaminated by industrial, residential and commercial activities. Three industrial parks and three residential sites were located in Kaohsiung's crowded Dashe, Renwu and Nanzih districts. The commercial sites were located at the crowded Houjing night market (Fig. 1). Here, the daily commercial use of plastics despite multiple government regulations for their control may contribute to elevated occurrence of organic contamination (Caliendo, 2015; Liao, 2017; Lin et al., 2017). To the best of our knowledge, this study is the first to investigate organic contamination caused by plastics used and collected from this typical popular and traditional commercial area in Asia. A detailed description of the sampling sites and the samples collected is provided in Table S1.

Sample collection and analysis basically followed the method described in Skrbic et al. (2016). Street dust was collected using a portable small-sized vacuum cleaner (8 BLACK + DECKER CHV1410L Cordless Dust-Buster Hand Vacuum). At each site, five sub-samples (within 50 × 50 m) were merged to form a composite sample (Skrbic et al., 2016). At least one duplicate sample was collected for each study area. Samples were kept in aluminum bags under cool conditions in a temperature-controlled box during the transportation to the laboratory. During the sampling and sample analysis, precautions were taken to avoid further PAE contamination in the laboratory (Lin et al., 2009). Samples were homogenously mixed and filtered through a stainless-steel sieve (60 mesh) and then stored at −20 °C until extraction.

2.2. Sample extraction and instrumental analysis

A PAE standard mixture containing dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (D*n*BP), benzyl butyl phthalate (BBP), di(2-ethylhexyl) phthalate (DEHP) and di-n-octyl phthalate (D*n*OP) was purchased from AccuStandard, Inc., New Haven, CT. The concentration of each PAE in the mixture was 1000 μg L^{-1}. Benzyl benzoate was used as an internal standard (99% purity; Sigma