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Carcinogenic profile, toxicity and source apportionment of polycyclic aromatic hydrocarbons accumulated from urban road dust in Tokyo, Japan

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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous in urban environments. Urban road dust (URD) generated by traffic is an important PAH accumulator. Twelve priority PAHs in < 2000 µm fraction of ten URD samples from Tokyo, Japan were characterized based on profile distributions, carcinogenicity, toxicity, and source apportionment by cluster analysis, biplot and diagnostic ratios. PAH concentrations (mg/kg dry weight) in arterial roads, highways, highway parking, highway drainage pit and residential area URD samples were 2.06–4.24, 0.25–3.37, 3.44, 4.94, and 5.26 respectively, dominated by the Σ4 rings (average 46%) and Σ5 + 6 rings (average 41%) PAHs. Biplot analysis revealed that the antecedent dry weather period, vehicle frequency and organic matter content were the dominant environmental factors governing PAH profiles of different road types. The total amount of carcinogenic PAHs in the residential URD (2.12 mg/kg) was higher than those in the arterial road (0.60–2.00 mg/kg) and highway (0.10–1.84 mg/kg) URD. Toxic equivalent concentrations (TECs) of residential, arterial road and highway URD were 0.54, (0.12–0.57), and (0.02–0.51) mg/kg, respectively. The dominant PAH sources were found to be petrogenic combustion in arterial road and highway URD, and pyrogenic combustion consisting of a mix of biomass, petroleum and traffic-related sources in the residential and highway drainage pit samples. This is also the first study to find that TEC-based toxicity should not be taken as a measure of URD toxicity.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are persistent and ubiquitous pollutants in urban environments. Some PAHs, such as benzo[*a*]pyrene, benz[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, chrysene, dibenz[*a*,*h*]anthracene, and indeno[1,2,3-cd]pyrene, have been reported to be toxic and mutagenic (Nisbet and LaGoy, 1992; USEPA, 1993; Petry et al., 1996), while benzo[*b*]fluoranthene has been reported to be a carcinogen precursor (Baird et al., 2005). PAHs are created by the incomplete combustion of the carbonaceous materials, such as fossil fuels, woodland, coal, or biomass, by lightening, industrial combustion, forest fires, traffic, or cigarettes. Thus, the main PAH sources in urban environments are automobiles and combustion by-products released by industrial and commercial activities (Mille et al., 2007; Dong and Lee, 2009).

Many PAHs are contained in urban road dust (URD), which is an important PAH accumulator and carrier in urban environments and is responsible for re-suspension and re-deposition of PAHs generated by traffic and other urban activities (Yu et al., 2014; Khanal et al., 2014). The PAHs attached to or combined with URD are dispersed, washed off, and built up by wind and surface runoff, and are considered to be one of the main sources of sediment contamination in urban water environments (Krein and Schorer, 2000; Aryal et al., 2005; Watanabe et al., 2011; Hiki et al., 2017).

In order to understand and mitigate the problem of PAHs in urban water environments, it is necessary to carry out source apportionment, by classifying them as pyrogenic, petrogenic or as a mixture of both (Yunker et al., 2002). Pyrogenic sources can be further divided into biomass and coal, and petrogenic sources into diesel, petrol, kerosene, and lubricating oil. PAH source apportionment can be done either according to the number of aromatic rings present or by calculating diagnostic ratios of PAH concentrations (Yunker et al., 2002; Pengchai

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St code	Coordinates	
St. 1	35^0 43’2”N	139^{0}45’57”E
St. 2	35^{0}42’27”N	139^{0}45’39”E
St. 3	35^{0}42’28-30”N	139^{0}46’15-12”E
St. 4	35^{0}38’36”N	139^{0}48’34-36”E
St. 5	35^{0}43’11”N	139^{0}45’54”E
St. 6	35^{0}31-40’01-30”N	139^{0}47-55’30-38”E
St. 7	35^{0}41’29-51”N	139^{0}47-52’23-47”E
St. 8	35^{0}31-33’01-13”N	139^{0}47’30-00”E
St. 9	35^{0}38’37-43”N	139^{0}51-53’58-51”E
St. 10	35^{0}31-40’01-30”N	139^{0}47-55’30-38”E

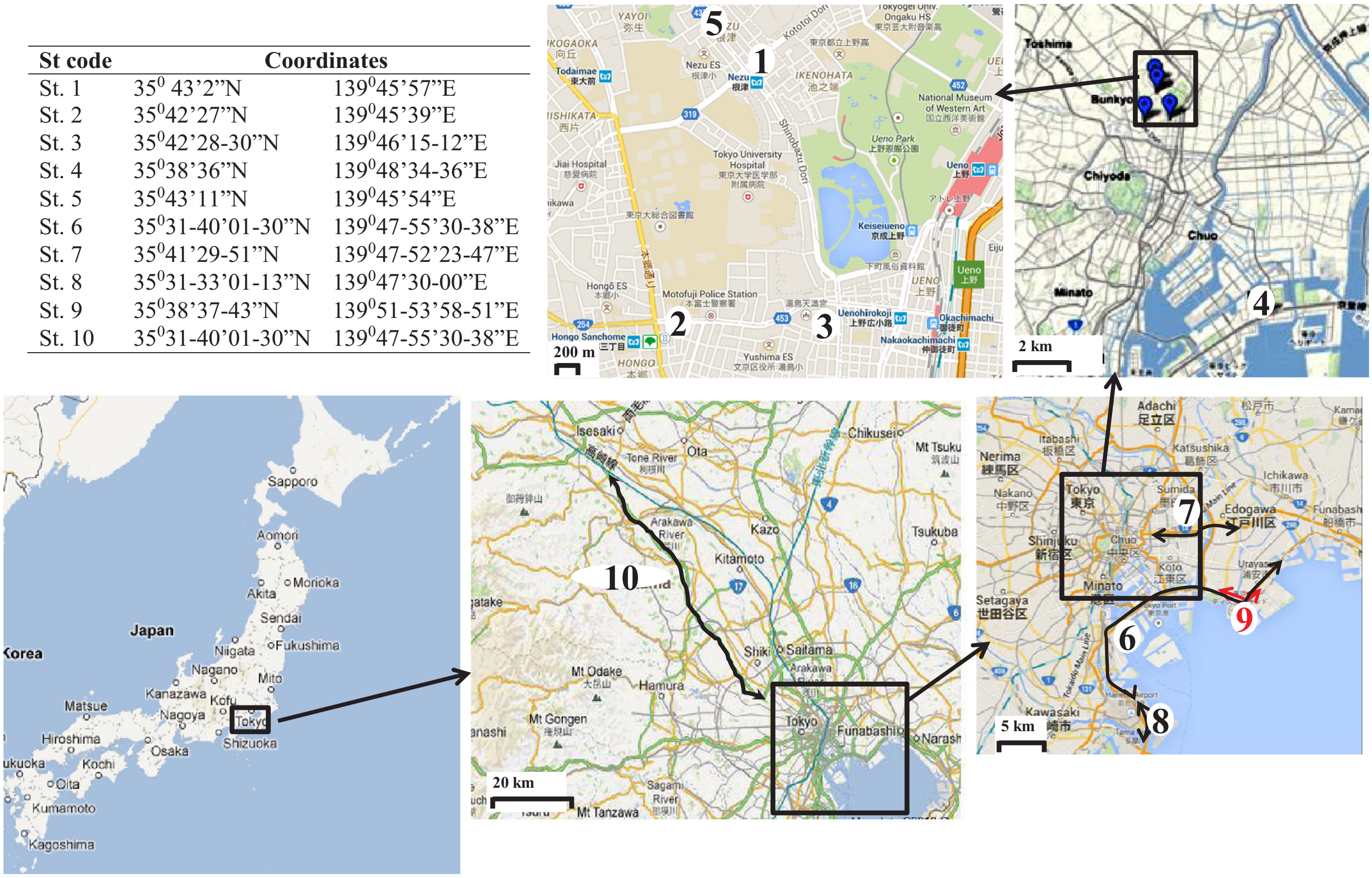

Fig. 1. Map of sampling sites with coordinates (adapted from google maps).

et al., 2004; Mille et al., 2007; Yu et al., 2014).

In general, PAHs with more than four aromatic rings are derived from pyrogenic sources and are believed to be more toxic than petrogenic PAHs, which have two or three aromatic rings (Wang et al., 2011). Understanding the sources and types of PAHs in a given urban environment would help to design appropriate mitigative measures. For example, as the number of carbons of aromatic rings, and hence the molecular weight increases, the aqueous solubility of PAHs decreases, resulting in such PAHs predominantly appearing in suspended solids and the sediment.

It should be noted, however, that not all PAHs are of environmental concern, as their toxicity, carcinogenicity, and mutagenicity vary by type (Nisbet and LaGoy, 1992; Petry et al., 1996). The bay-region theory, DNA-binding intermediates, and metabolically-induced mutations in oncogenes are the most widely-accepted theories of carcinogenic potential of PAHs (Yan, 1985; Baird et al., 2005; Vijayalakshmi and Suresh, 2008). Since, complex mixtures of PAHs occur in URD, it is difficult to assess the carcinogenicity and toxicity of any individual PAH, as combinations of two or more PAHs may exhibit different carcinogenic or toxic effects then they would when considered individually. Hence, the concepts of cumulative carcinogenic potential, toxic equivalent concentration, toxic units and source apportionment are vital when characterizing the PAHs in URD (Nisbet and LaGoy, 1992; Petry et al., 1996; Khanal et al., 2014).

Various researchers have analyzed the PAH distribution in URD from around the world, such as in Tokyo, Japan (Murakami et al., 2005), Dalian, China (Wang et al., 2009), Cairo, Egypt (Hassanien and Abdel-Latif, 2008), Ulsan, South Korea (Lee and Dong, 2010), Guangzhou, China (Wang et al., 2011), Sydney, Australia (Nguyen et al., 2014), and Isfahan, Iran (Soltani et al., 2015). Most of these studies have focused on chemical characterization, based on size fractions, seasonal variations, pavement types, traffic volumes, vehicle speeds, and other aspects of the urban environments.

In addition, the toxicity, calculated in terms of the toxic equivalent concentration (TEC) (Petry et al., 1996) has been proposed as a direct measure of URD toxicity. It has been reported in the literature that higher PAH concentrations lead to higher TECs and hence higher toxicity, as the TEC is calculated by assigning a toxicity factor to each PAH and summing these toxicity values (Dong and Lee, 2009; Lee and Dong, 2010; Nguyen et al., 2014). However, Khanal et al. (2014) found there was no significant correlation between the PAH concentration in URD and toxicity to Ostracod. The general presumption of taking URD toxicity to be positively correlated with PAH concentration and TEC must therefore be validated by actual toxicity data, taking account of the environmental variables related to the PAH source and distribution.

In a highly urbanized areas with heavy traffic, the lack of clear data on the distribution of PAH in URD, their sources, and their toxicity, has made it very difficult for decision makers to propose ways of combating PAH pollution. In addition, since URD is a non-point pollution source, it is very difficult to set standards to control PAH levels in urban environments. There are also very limited studies on assessing the environmental factors that govern PAH profile distribution, or integrated studies of its carcinogenicity, toxicity, and source apportionment based on URD. Hence, the main objective of this research was to characterize the profile distributions, carcinogenicity, toxicity, and source apportionment of PAHs. Furthermore, the diagnostic ratio of individual PAHs isomers was also analyzed to apportion the PAH sources originating from residential area, arterial roads, highways, highway parking, and highway drainage pit.