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Status, source and health risk assessment of polycyclic aromatic hydrocarbons in street dust of an industrial city, NW China

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ABSTRACT

The status, source and health risk of street-dust-borne polycyclic aromatic hydrocarbons (PAHs) in Lanzhou of Northwest China were investigated. The total level of the 21 PAHs ranged from 1470 to 13,700 μg kg^{-1} and that of the 16 priority PAHs from 1240 to 10,700 μg kg^{-1}. Higher levels of PAHs were mainly distributed in the Chengguan and Qilihe districts at Lanzhou central areas, and the lower levels were in Anning and Xigu districts. The level of seven potential carcinogenic PAHs generally accounted for 35–40 percent of total PAHs, and the PAHs contained two to four rings, mainly phenanthrene, benzo[*b*] fluoranthene and fluoranthene. The total level of PAHs increased with the decreasing particle size in the street dust. The correlation analysis suggested that the total organic carbon (TOC) was only slightly affected the PAH accumulation in street dust. The isomer ratios and principal component analysis indicated that the dust-borne PAHs in the dust were derived primarily from the combustion of biomass, coal and petroleum emission. The toxic equivalent concentrations (BaP_{eq}) of dust-borne PAHs ranged from 115 to 827 μg BaP_{eq} kg^{-1}, with a mean of 300 μg BaP_{eq} kg^{-1}. The 95 percent upper confidence limit of Incremental Lifetime Cancer Risk due to human exposure to urban surface dust-borne PAHs in Lanzhou urban area was 2.031×10^{-6} for children and 1.935×10^{-6} for adults.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) as ubiquitous environmental pollutants primarily result from the incomplete combustion of predominant anthropogenic sources, especially fossil fuel, biomass, and coal. PAHs are receiving extensive attention because of their adverse effects on human health including high toxicity, mutagenicity and carcinogenicity. Sixteen parent PAHs have been identified by the United States Environment Protection Agency (US EPA) as priority pollutants, and among them, the potential carcinogenic PAHs include benzo[a]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*a*] pyrene, benzo[*k*]fluoranthene, indeno[1,2,3-cd]pyrene and dibenz[*a*,*h*] anthracene. Furthermore, PAHs are considered as persistent organic pollutant (POP) candidates that merit further investigation for possible early inclusion into the Stockholm Convention on POPs (World Wide Found (WWF), 2005). Therefore, PAHs pollution has attracted growing attention recently, and numerous investigations show that PAHs are ubiquitous in various environmental media (Chung et al., 2007; Jiang et al., 2009; Lorenzi et al., 2011; Choi et al., 2012; Krugly et al., 2014).

Street dust in urban areas is an indicator of toxic pollutants deposited from the atmosphere (Tsai et al., 2004; Wu et al., 2005; Wang et al., 2011). Street dust consists of vehicle exhaust, airborne sinking particles in air, house dust, soil dust and air- and wind-borne aerosols (Liu et al., 2007; Martuzevicius et al., 2011; Choi et al., 2012), and significantly contributes to urban pollution. PAHs can accumulate in street dust via atmospheric deposition by sedimentation interception and may threaten human health if reaching the levels of toxic pollutants (Dong and Lee, 2009; Lorenzi et al., 2011; Wang et al., 2011; Choi et al., 2012). The tightly-packed buildings along with the urban expansion limit air circulation and thus lead to enhanced PAHs accumulation in street dust (Kong et al., 2012; Li et al., 2014). Multiple-source materials especially pyrogenic and petrogenic sources contribute to PAH accumulation in street dust. Street dust which contains a complex mixture of petrogenic and pyrogenic PAHs is a key non-point source of PAHs (Boonyatumanond et al., 2007; Liu et al., 2007). The PAH-polluted street dust presents higher health risk to children and adults compared with automobile emissions (Wang et al., 2011; Krugly et al., 2014). Street dust is chemically similar to the primary portion of atmospheric aerosols in some respects, and is

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indeed dynamically related to atmospheric aerosols through re-suspension into and re-deposition from the atmosphere (Rogge et al., 1993; Liu et al., 2007; Li et al., 2014). However, there are few studies about PAHs in street dusts.

Lanzhou as a rapidly developing city was chosen in this study, because this region is considered as a major work place in the northwestern China. Lanzhou contains many workplace segments such as petrochemical complex, smelters, steel and non-steel industries, construction materials, and chemical plants, etc. Lanzhou with heavy automobile traffics and large petroleum plants is also one densely-populated large city in northwest China. The main fuel in its energy consumption structure is coal, causing serious air pollution in this area. Moreover, Lanzhou is located in a valley with a typical Canyon topography between the northern and southern hills, and thus the specific topography there resembles that of a mountain-surrounded valley that traps the dirty air within. As a result, the poor air quality in the Lanzhou due to the high-level total suspended particles (TSP) and photochemical smog (Tang et al., 1985; Jiang et al., 2001; Ta et al., 2004; Gao et al., 2007; Chu et al., 2008) results in frequent occurrence of respiratory diseases, such as asthma and lung cancer (Gao et al., 2007; Pan et al., 2010; Tao et al., 2012; Chen et al., 2013). However, little is known about the PAH pollution in street dust of Lanzhou. The purposes of the present study are to: (1) determine the level and distribution of PAHs pollution, (2) elucidate the potential input sources, and (3) assess the potential health risks of PAHs in Lanzhou urban street dust.

2. Materials and methods

2.1. Sampling area description

Lanzhou (35°34′20″–37°07′07″N, 102°35′58″–104°34′29″E) with a total area of 688.9 km^2 is the capital of Gansu Province in Northwest China. It has a continental climate of the North Temperate Zone, with an annual average temperature of 9.5 °C, relative humidity of 57 percent, and annual rainfall of 327.7 mm. About 50 percent of the annual precipitation (327.7 mm) is centralized during the July-September period. Under the topographic effects, the monthly average of the surface wind speed is approximately 0.8 m/s (Ta et al., 2004). About 2.5 million people live in the four urban districts, including Chengguan district (CG), Qilihe district (QLH), Anning district (AN), and Xigu district (XG). Among them, both Chengguan district and Qilihe district have longer history of living and lager population of inhabitants. The average traffic volumes in Chengguan and Qilihe are 2600 and 2333 vehicles per 24 h, respectively. Chengguan is a residential-commercial mixed centre of Lanzhou. Anning developed as a suburban area in recent years, with an average traffic volume of 1206 vehicles per 24 h. Xigu has been an industrial estate of organic pollution for decades (Tang et al., 1985), with an average traffic volume of 1633 vehicles per 24 h.

2.2. Dust sampling and preparation

The PAH pollution in urban Lanzhou was assessed using a stratified sampling strategy and a total of 32 street dust samples were collected in October 2011. The sampling sites covered the four urban districts, including Chengguang: CG1–CG8, Anning: AN1–AN8, Qilihe: QLH1–QLH8, and Xigu: XG1–XG8 (Fig. 1). At each site, street dusts were collected from within 2 m^2 on the road, using polyethylene brush, tray and containers, and samples were collected in polyethylene containers. The samples collected at AN2, AN4, CG1, CG5, QLH3, QLH5, XG1 and XG8 were selected randomly and divided into four categories depending on particle size measured by laboratory test sieves: <100 μm, 101–500 μm, 501–1000 μm, and >1000 μm. The samples were air-dried at room temperature and crushed after removing stones to pass a 100-mesh sieve, and then stored in a refrigerator until analysis. Dust samples (each 1 g) were isolated for measurement of percentage moisture and total organic carbon (TOC). Dust moisture contents were measured by drying at 105 °C to a constant weight. After that, these samples were put into a muffle furnace for TOC detection by measuring their loss upon ignition at 550 °C.

2.3. Reagents and glassware

A composite standard solution with 18 PAHs including naphthalene (Nap), 2-methylnaphthalene (2-MNa), 1-methylnaphthalene (1-MNa), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fl), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benz[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[1,2,3-cd]pyrene (InP), dibenz[*a,h*]anthracene (DBA), and benzo[*g,h,i*]perylene (BP) each at a concentration of 2000 μg mL^{-1}, and a deuterated PAH mixture standard solution containing d_8-Nap, d_{10}-Ace, d_{10}-Phe, d_{12}-Chr, d_{12}-DBA, and d_{12}-Pyr each at a concentration of 2000 μg mL^{-1} were purchased from Supelco (Bellefonte, PA, USA). Individual solutions of benzo[e]pyrene (BeP), Retene (Ret), perylene (Per) and coronene (Cor) at 200 μg mL^{-1} were also obtained from Supelco (Bellefonte, PA, USA). A working standard solution containing 21 native PAHs and 6 deuterated PAHs was prepared with isooctane before use. Silica gel (100–200 mesh, Qingdao Haiyang Chemical Co., Shandong, China) was activated for about 16 h at 130 °C and granular anhydrous sodium sulfate was baked at 450 °C for 5 h before use. All solvents and chemicals were of analytical grade and redistilled before use.

2.4. Sample extraction, cleanup and analysis

Each sample (10 g) spiked with surrogates (d_8-Nap, d_{10}-Acy, d_{10}-Phe, d_{12}-Chr, d_{12}-Pyr and d_{12}-DBA) was mixed with 10 g of anhydrous sodium sulfate and then Soxhlet-extracted with 200 mL of hexane/acetone (1:1 *v/v*) for 36 h. The extracts were concentrated by rotary vacuum evaporation and solvent-exchanged to hexane. The concentrated extracts were cleaned up by a silica gel column chromatography (25 cm × 1 cm i.d). The glass chromatographic column, fitted with a Teflon stopcock, was packed from bottom to top with glass wool, 10 g of activated silica, and 2 g of anhydrous sodium sulfate. After introduction of the extract, the first fraction eluted with 25 mL of hexane was discarded, while the second PAH-containing fraction eluted with 35 mL of *n*-hexane/dichloromethane (3:2 *v/v*) was collected. The eluate was concentrated to 1 mL and solvent-changed to isooctane, and then further concentrated to 0.2 mL under a gentle stream of nitrogen before gas chromatography/mass spectrometry (GC/MS) analysis.

The PAHs were detected on an Agilent 6890 gas chromatograph-5975 mass selective detector (GC–MS) equipped with a DB-5 column (30 m × 0.25 mm i.d., 0.25 μm film thickness), using helium as the carrier gas. The oven temperature program was set as follows: initially at 60 °C with retention for 2 min, heated at 5 °C min^{-1} to 190 °C, and then at 10 °C min^{-1} to 290 °C. The injector temperature was 280 °C. The MS was operated in the electron impact ionization mode with electron energy of 70 eV and the mass range scanning was from 50 to 550 amu under the selected ion monitoring (SIM) mode. The ion source, quadrupole and transfer line were held at 230, 150 and 280 °C, respectively. The sample extracts (each 1 μL) were injected in the splitless mode. The individual PAHs were identified on basis of the selected ions and comparison of retention time between samples and the standard solution. The individual PAHs were quantified using internal calibration.

2.5. Risk assessment

The exposure risk of environmental PAHs was quantified using incremental lifetime cancer risk (ILCR) based on the U.S. EPA standard models (US EPA, 1991; Chen and Liao, 2006; Peng et al., 2011; Wang et al., 2011). The ILCRs in terms of ingestion, dermal contact and inhalation after exposure to urban surface dust-borne PAHs in different areas of Lanzhou were calculated as follows:

$$ILCRs_{Ingestion} = \frac{CS \times (CSF_{Ingestion} \times \sqrt[3]{(BW/70)}) \times IR_{Ingestion} \times EF \times ED}{BW \times AT \times 10^6}$$

$$ILCRs_{Dermal} = \frac{CS \times (CSF_{Dermal} \times \sqrt[3]{(BW/70)}) \times SA \times AF \times ABS \times EF \times ED}{BW \times AT \times PEF}$$

$$ILCRs_{Inhalation} = \frac{CS \times (CSF_{Inhalation} \times \sqrt[3]{(BW/70)}) \times IR_{Inhalation} \times EF \times ED}{BW \times AT \times PEF}$$

where CS is the sum of converted PAH levels based on toxic equivalents of BaP using the toxic equivalency factor (TEF). CSF is carcinogenic slope factor (mg kg^{-1} $day^{-1})^{-1}$, BW is body weight (kg), AT is the average life span (years), EF is the exposure frequency (day $year^{-1}$), ED is the exposure duration (years), $IR_{Inhalation}$ is the inhalation rate (m^3 day^{-1}), $IR_{Ingestion}$ is the soil intake rate (mg day^{-1}), SA is the dermal surface exposure (cm^2), AF is the dermal adherence factor (mg cm^{-2} h^{-1}), ABS is the dermal adsorption fraction, and PEF is the particle emission factor (m^3 kg^{-1}). PEF is particle emission factor (m^3 kg^{-1}); $CSF_{Ingestion}$, CSF_{Dermal} and $CSF_{Inhalation}$ of BaP were addressed as 7.3, 25, and 3.85 (mg kg^{-1} $day^{-1})^{-1}$, respectively, determined by the cancer-causing ability of BaP (Peng et al., 2011). All the parameters used in these models for children (1 to 6 years old) and adults (7 to 31 years old) were based on the Risk Assessment Guidance of U.S. EPA and related publications (US EPA, 1991; Wang et al., 2011).

2.6. Quality assurance/quality control

The limit of detection (LOD) for individual PAHs ranged from 0.15 to 0.41 μg kg^{-1} with a signal-to-noise ratio of 3:1 in the blank sample ($n=7$). The spike blanks, solvent blank and duplicate samples were analyzed each in sextuplicate, and no interferences were detected. The procedure was also checked