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Polycyclic aromatic hydrocarbons in the surface soils from outskirts of Beijing, China

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Abstract

Surface soils from the outskirts of Beijing were analyzed for 16 priority polycyclic aromatic hydrocarbons (PAHs) using gas chromatography and mass spectrometry (GC–MS). The distribution map of total PAHs content was obtained as a contour plot. The concentration range of 16 PAHs varied by over two orders of magnitude from 0.016 μg g^{-1} in rural to 3.884 μg g^{-1} in suburban soils with the relatively standard deviation of 70.5%, showing large differences in the extent of PAHs pollution at the various sampling sites. It was notable that the concentration of BaP was 0.005–0.270 μg g^{-1} with a mean of 0.055 μg g^{-1}. In general, the distribution of PAHs centered on the high molecular weight PAHs known to be carcinogenic. The 4–6 ring PAHs represented about 66% in rural samples and 70% in suburban soils of the total PAHs found. There was relatively good relationship among most of the individual PAHs and the compounds of Pyr, BaA, Flu, BbF, BaP, Chr and Ph gave strong correlation ($r > 0.8$) with the sum of PAHs. The selected marked compounds, a principal component analysis (PCA) and special PAHs compound ratios (Ph/An vs Flu/Pyr; $\sum \mathrm{COMB}/\sum \mathrm{EPA}-\mathrm{PAHs}$) suggest the pyrogenic origins, especially traffic exhausts, are the dominant sources of PAHs in Beijing outskirts soils.

Keywords: PAHs; Surface soils; Principal component analysis; Traffic emission; Beijing

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1. Introduction

There is a globally increasing concern about environmental pollution by persistent organic pollutants (POPs). Besides 12 POPs listed in "Stockholm Convention", the polycyclic aromatic hydrocarbons (PAHs) were included among 16 POPs designated in "Convention of Long-range Transboundary Air Pollution" adopted by United Nations Economic Commission for Europe. PAHs are also identified as high priority chemicals in "Regionally-based assessment of Persistent Toxic Substances" in Region VII (including China) conducted by United Nations Environment Programme and Global Environmental Fund. As ubiquitous contaminants, PAHs are particularly harmful due to their carcinogenic and mutagenic properties (IARC, 1983; Perera, 1997). The US Environmental Protection Agency (EPA) has promulgated 16 unsubstituted PAHs (EPA-PAH) as priority pollutants. Eight PAHs (Car-PAHs) typically considered as possible carcinogens are: benz(*a*)anthracene, chrysene, benzo(*b*)fluoranthene, benzo(*k*)fluoranthene,

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benzo(*a*)pyrene, dibenz(*a,h*)anthracene, indeno(1,2,3-*cd*)pyrene and benzo(*g,h,i*)perylene (Menzie et al., 1992). Although natural sources affect the concentrations in local areas, PAHs occur primarily due to the anthropogenic inputs including the incomplete combustion or pyrolysis of organic material and the direct release of oil or its products. The previous investigations showed traffic was the major PAHs source in metropolitan areas (Rogge et al., 1993a; Van Metre et al., 2000; Okuda et al., 2002). Comparatively stable molecular structure, slow rates of photochemical decomposition and biodegradation contribute to the persistence of PAHs in the environment. Soil system seems to be the important long-term repository for PAHs and is considered to be a steady indicator of the environmental pollution state (Wild et al., 1990; Wild and Jones, 1995). Accumulation of PAHs in soils may lead to further potential contamination of vegetables and food chains (Meharg et al., 1998; Kipopoulou et al., 1999), and then cause direct or indirect exposure to human. Moreover, leaching, evaporation and migration are possible PAHs sources of atmospheric or groundwater contamination (Bispo et al., 1999; Cousins et al., 1999). Therefore, knowledge of soil contamination with PAHs is needed to avoid any food production risk and to restrict the deleterious effect of these contaminants. Many studies have been reported for the PAHs in soils in some countries (Edwards, 1983; Jones et al., 1989; Wilcke et al., 1999; Wilcke, 2000). However, in China most investigation only directed towards determining PAHs concentrations and distributions in water (Bi et al., 2001; Wang et al., 2004), air (Katagiri et al., 1996; Li et al., 2001) and sediment (Liu et al., 2000; Yang, 2000; Wu et al., 2003). Up to now, comparatively few investigations have been conducted to address the accumulation and distribution of PAHs in the soils (Chu et al., 2003; Wang et al., 2003; Tao et al., 2004).

Beijing (39°48′N and 116°28′E), as the capital, is the second largest city in China. The high population (over 13 million) and rapid industrialization during the last decades have resulted in a lot of environmental problems. The vehicle in Beijing grows at a rate of 15% per year through the 1990s, and now exceeds 2.0 million (He et al., 2001). The PAHs concentrations calculated in terms of total suspended particles were 178 $\mu g\ g^{-1}$ in summer and 749 $\mu g\ g^{-1}$ in winter (Okuda et al., 2002). The benzo(*a*)pyrene concentration in outdoor air in residential areas of Beijing was approximately 15 times higher than that in residential areas around a main road of Tokyo (Katagiri et al., 1996). Therefore, it is urgent to investigate PAHs in the environment of Beijing. By now, scarce research concerned about PAHs in the Beijing soils (Chu et al., 2003; Ma et al., 2003).

In 2001, the Chinese government carried out an investigation to assess the state of soil contamination on the Beijing outskirts where are undergoing great changes due to the rapid urbanization and industrial development. This paper described here is part of the wider research project. In the present study, surface soils from Beijing rural and suburban areas were investigated to identify the current PAHs contamination status, composition and possible sources.

2. Material and methods

2.1. Materials and reagents

The standard solutions including 16 EPA-PAHs (naphthalene, Na; acenaphthylene, Acy; acenaphthene, Ace; fluorene, Fl; phenanthrene, Ph; anthracene, An; 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; benzo(*g,h,i*)perylene, BghiP) each at the concentration of 1000 $\mu g\ ml^{-1}$ were obtained from Supelco (Bellefonte, PA, USA). 2-flurobiphenyl was purchased from Aldrich chemical company. The standards were further diluted with isooctane to prepare working standards. Silica gel (100–200 mesh; Qingdao Haiyang Chemical Company, China) was activated at 130 °C for 12 h. All the solvents (Beijing Chemical Factory, China) were of analytical purity and redistilled in all-glass system prior to use.

2.2. Sampling and preparation

Surface soil samples at the depth about 5–30 cm were collected covering an areas of about 1000 km^2 in April 2001 and the sampling sites are shown in Fig. 1. Samples 1–14 were from rural area and samples 15–47 were from suburban area.

Firstly, multiple cores covering about 1 km^2 were collected to get one subsample with stainless steel drill and then 16 vicinal subsamples were mixed fully to obtain one pooled sample representing 16 km^2 areas. The soil was air-drying in fume hood at room temperature, sieved to <1 mm and stored in glass bottles at −4 °C. The remaining water content in the soil was determined gravimetrically after drying individual sample in an oven at 105 °C for 12 h. All results were reported as dried weight basis.

2.3. Sample extraction and clean-up

After 1 ml of 2-flurobiphenyl (4.0 $\mu g\ ml^{-1}$) added as surrogate, accurately weighed soil (about 5 g) was extracted by ultrasonic in 30 ml acetone/petroleum ether (1/1) thrice during 15 min followed and separated by centrifugation. The solvent was evaporated by K-D apparatus and then reduced to about 1 ml by a gentle stream of nitrogen.