# **Polycyclic Aromatic Hydrocarbons in Urban Street Dust and Surface Soil: Comparisons of Concentration, Profile, and Source**

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Abstract Street dust and surface soil samples in urban areas of Dalian, a coastal city in Liaoning Province, China, were collected and analyzed for 25 polycyclic aromatic hydrocarbons (PAHs). The concentrations, distribution, and sources of PAHs in dust and soil were determined. The concentrations of total PAHs in street dust ranged between 1890 and 17,070 ng/g (dry weight), with an average of 7460 ng/g, whereas the concentrations of total PAHs in surface soil varied greatly, from 650 to 28,900 ng/g, with a mean value of 6440 ng/g. Statistical paired t-test confirmed that total PAH concentrations have no significant difference between street dust and surface soil. Mean PAH concentrations in two type samples were much higher at industrial sites than at business/residential or garden sites. PAHs were dominated by higher molecular weight PAH (4- to 6-ring) homologues, which accounted for about 73% and 72% of total PAHs in street dust and surface soil, respectively. Principal component analysis was used in source apportionment of PAHs in dust and soil. Pyrogenic and petrogenic sources contributed 70% and 22.4% of total PAHs in street dusts, and fossil fuel (coal and petroleum) and biomass combustion accounted for 64.4% and 5.6% of total PAHs in pyrogenic sources, respectively. In surface soil, total PAHs were dominated by pyrogenic sources. The diagnostic ratios of benz[a]anthracene/chrysene confirmed

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Y.-F. Li (⊠) Science and Technology Branch, Environment Canada, Toronto, ON, Canada e-mail: yi-fan.li@ec.gc.ca that PAHs in street dust and surface soil of a Dalian urban zone might come mostly from the emission of local sources.

Polycyclic aromatic hydrocarbons (PAHs) are organic chemicals composed of fused benzene rings, which are highly persistent, toxic, and widespread environmental contaminants. Natural and anthropogenic processes generate PAHs during the incomplete combustion of all types of organic matters. PAHs are emitted to ambient air as particles and gases. Atmospheric PAHs that escape photolysis and radical oxidative degradation in the air are accumulated to land surfaces through dry and wet deposition. Once deposited, PAHs tend to accumulate in soil or to become roof and street dust in the urban environment for a long period of time (Ockenden et al. 2003). Atmospheric deposition is a major source for PAHs in surface soil and street dust (Takada H et al. 1990; Wild and Jones 1995; Murakami et al. 2005). In addition, surface soil and street dust PAHs will go back to the atmosphere by wind raise and volatilization (Manoli et al. 2002, 2004). Moreover, street dust and surface soil contained PAHs are carried into the drainage network by storm runoff and are eventually transported to aquatic environments during rain season (Mai et al. 2003; Boonyatumanond et al. 2006). Therefore, soil and dust are natural sinks and environmental reservoirs for PAHs in the urban environment. Accumulation of PAHs in dust and soil may lead to contamination of vegetables and food chains, leading to direct or indirect human exposure (Kipopoulou et al. 1999; Tao et al. 2004; Zohair et al. 2006; Cai et al. 2007). Thus understanding of the fate of PAHs deposited to the street dust and surface soil is very important to evaluate the risk of human exposure and environmental contamination.

Soil as a repository of all types of chemical inputs, is important to the sources and pools of PAHs on the regional scale (Wild and Jones 1995). A number of studies have been conducted on the levels, distributions and sources of PAHs in urban soil (Zhang et al. 2006; Morillo et al. 2007; Wilcke 2007; Zuo et al. 2007). The mean PAH levels varied largely in different cities and the pyrogenic origins, especially motor vehicle exhausts, are the major sources of PAHs in urban soils (Zhang et al. 2006; Morillo et al. 2007).

Apart from the widely investigated PAHs in urban surface soil, a few studies have reported PAHs in street dusts previously (Takada et al. 1990; Boonyatumanond et al. 2007; Liu et al. 2007). Street dust is chemically similar to the primary portion of atmospheric aerosol in some respects and indeed bears a dynamic relationship to atmospheric aerosol through resuspension into and redeposition from the atmosphere (Rogge et al. 1993). Multiple-source materials contribute to PAHs in street dust, and street dust PAHs mainly came from the mixing of pyrogenic and petrogenic sources. Street dust is a complex mixture of petrogenic and pyrogenic PAHs and is an important nonpoint source (Takada et al. 1990; Boonyatumanond et al. 2007; Liu et al. 2007).

A number of studies have been conducted on the pollution of PAHs in surface soil and street dusts. However, relatively few studies have explored the PAH contamination relationship in soil versus dust. It is necessary to investigate the contamination of PAHs in surface soil and street dust and to identify the pattern of PAH profiles in urban areas. The main objectives of this study were (1) to determine the distribution and concentrations of PAHs in soil and dust of urban areas, (2) to compare the PAH concentrations in street dust and surface soil, and (3) to identify and compare the sources of PAHs in street dust and surface soil. Dalian, as a typical coastal city situated in the northeastern monsoonal area of China (E120°58′– 123°31′, N38°43′–40°10′), was selected as the urban area for this study.

# **Materials and Methods**

# Sampling

The concurrent sampling of surface soil and street dust was performed in the urban areas of Dalian (Fig. 1) during July 2007. In total, 13 surface soil and street dust samples were collected in pairs, using a metal tool. These sampling sites include four industrial sites (I1, I2, I3, and I4), seven business/residential sites (B/R1, B/R2, B/R3, B/R4, B/R5, B/R6, and B/R7), and two garden sites (G1 and G2). In soil sampling places a field of about  $100 \times 100 \text{ m}^2$  was



**Fig. 1** Map of the surface soil and street dust sampling sites in Dalian. The distances of soil and dust sampling sites ranged from 100 to 1000 m, with an average of about 500 m

selected and samples were collected at the four corners and the center of sampling fields. An approximately 0.5-kg soil sample was taken from the top 5 cm of the soil after removal of the large stones and pieces of vegetation. In the immediate vicinity of each soil sampling site, a street was selected as the street dust sampling place. The distances of soil and dust sampling sites ranged from 100 to 1000 m, with an average of about 500 m. Street dust samples were collected by sweeping the street surface with a brush and collecting the dust on a piece of aluminum foil. Each soil and dust sample consisted of five subsamples collected from the surroundings of each site. Samples were packed in a solvent-rinsed glass bottle with a Teflon-lined cap, sent to the laboratory, and stored at  $-20^{\circ}$ C until required for extraction.

# Dust and Soil Characterization

Five grams of soil was isolated for percentage moisture and total organic carbon (TOC) determination. Dust and soil moisture contents were determined by drying at 105°C to a constant weight. After determination of moisture contents, these soils were put into a muffle furnace for TOC determination by measuring their loss upon ignition at 550°C. The moisture contents ranged from 4.93% to 4.98% for dust and from 3.19% to 10.99% for soil, respectively. TOC contents varied between 3.31% and 11.36% for dust and between 1.42% and 12.88% for soil.

#### Extraction and Analysis

Samples were treated, extracted, and analyzed according to the methods established at the National Laboratory for Environmental Testing (NLET), Environment Canada. All solvents used were of pesticide-grade purity (J.T. Baker, USA). Twenty-five PAH standards were purchased from

Supelco, Inc. (USA). Two-ring PAHs are indene (Ind), tetrahydronaphthalene (ThN), naphthalene (Nap), 2-methynaphthalene (2-MN), and 1-methynaphthalene (1-MN). Three-ring PAHs are acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fl), phenanthrene (Phe), anthracene (Ant), dibenzothiopene (Dib), and retene (Ret). Four-ring PAHs are fluoranthene (Flu), pyrene (Pyr), benz[a]anthracene(BaA), and chrysene(Chr). Five-ring PAHs are benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[e]Pyrene (BeP), benzo[a]pyrene (BaP), perylene (Per), and dibenz[a,h]anthracene (DaA). Six-ring PAHs are indeno[1,2,3-cd] pyrene (InP) and benzo[g,h,i]perylene (BghiP). Twenty grams of each soil sample was accurately measured into a precleaned extraction thimble and spiked with a recovery standard containing a surrogate standard mixture (naphthalene-d10, fluorene-d10, pyrened10, and perylene-D12). To all the extracts, an appropriate volume of PAH surrogate standard mixture was added.

The soil and dust samples were then Soxhlet extracted for 24 h with a 100-ml mix of solvent (n-hexane/acetone; 1:1, v/v). The extract was filtered through a funnel filled with anhydrous sodium sulfate, then rotary-evaporated to 1 mL. The extract was passed through a 10-g silica gel column (Silica 60; Merck, Germany), eluted with a 60-mL mix of hexane and dichloromethane (1:1, v/v). The elution was rotary-evaporated to 2 mL and then reduced to 1 mL under a gentle nitrogen gas flow. All PAHs were identified and quantified by GC-MS (Finnigan PolarisQ). The mass spectrometer was operated in electron impact mode (70 eV). Helium gas was used as a carrier. The GC column was an HP-5MS fused silica column (60 m  $\times$  0.25-mm i.d., 0.25-µm film thickness). The column oven temperature was programmed at a rate of 25°C/min from an initial temperature of 60°C to a temperature of 180°C (1-min hold), then at 3°C/min to 280°C (30-min hold). Injector, transfer line, and ion source temperatures were 280, 250,

Fig. 2 Selected ion

PAHs

94% for pervlene-D12 in all samples. Spike samples were included at a rate of one for every 10 soils extracted, and the recoveries of all 25 PAHs varied from 78% to 110%. The method detection limit (MDL) for the 25 PAHs ranges from 0.063 to 7.991 ng/g dw (dry weight), with a mean value of 1.362 ng/g dw.

# **Results and Discussion**

deuterated PAHs.

Quality Assurance/Quality Control

#### General PAH Concentration Trends

Concentrations of total PAHs in street dust and surface soil for the 13 sites are shown in Fig. 3. In general, the concentrations of total PAHs in street dust ranged between 1894 and 17,074 ng/g dw, with an average of 7458 ng/g, whereas the concentrations of total PAHs in surface soil varied greatly, from 649 to 28,904 ng/g, with a mean value of 6,439 ng/g. The lowest and the highest concentrations in dust samples were found at Sites G1 and I3, which are a garden site and an industrial site, respectively. The PAH congeners in street dusts exhibited a rather uniform distribution of concentrations, with less than one order of magnitude of variation. For surface soil, the lowest and the highest concentrations were measured in samples from

and 250°C, respectively. PAHs were monitored in selected

ion monitoring (SIM) mode. Figure 2 presents the GC-MS

chromatogram for the mixture of the 25 PAHs and 4

All samples were spiked with a labeled recovery standard (naphthalene-d10, fluorene-d10, pyrene-d10, and perylene-

d12) prior to extraction. The average surrogate standard

recoveries ranged between 75% for naphthalene-D10 and



**Fig. 3** Concentration levels of total PAHs in surface soil and street dust for the 13 sampling sites in the city of Dalian



Sites B/R4 and I3, which are a business/residence site and an industrial site, respectively. In comparison to the dust samples, the total PAH levels in soil varied more and the variation was more than one order of magnitude.

Variations of PAH concentrations in dust and soil were also observed in different areas within Dalian. Table 1 reports the concentrations of total PAHs in three regions: industrial area, business/residential area, and gardens. In street dust, the highest mean concentration (12,266 ng/g) appeared at industrial sites, followed by business/residence (5830 ng/g) and garden (3540 ng/g) sites. In surface soil, the concentration order was industrial sites (13,068 ng/g) > garden sites (4391 ng/g) > business/residence sites (3237 ng/g). It is not unexpected that concentrations of PAHs were much higher at industrial sites than at business/residential or garden sites for both soil and dust samples due to the significant contributions of industrial production to PAH

It is commonly observed that PAH concentrations in environmental media are affected by TOC content, with higher concentrations accompanied by high TOCs (Wilcke 2000). Relationships between PAH concentrations and TOC contents of soil and dust were examined by correlation analysis (Fig. 4). There was a strongly significant positive correlation (R = 0.82, p < 0.001) between total PAH concentrations and TOC content in soil, whereas no correlation (R = 0.31, p = 0.30) was found for total PAHs in dust. It is evident that the TOC influenced the distribution of PAHs in surface soil but did not influence the distribution of PAHs in street dust. The possible reason is that soil and dust have different physical and chemical components, which induce different organic matter distributions. The main organic matters in soil are humic substances and they are homogeneous (Senesi 1992). Dust is a mixture of atmospheric aerosol, sands, asphalt, tire, and soil participles and the origins of organic matters are complex (Takada et al. 1990). Therefore, organic matters in dust are less homogeneous than in soil, suggesting that TOC has less influence on PAH distribution in dust than in soil.

# Relationship of PAH Concentrations in Samples of Street Dust versus Surface Soil

Total PAH levels in surface soil samples ( $C_S$ ) were compared with those in street dust samples ( $C_D$ ) to evaluate their relationship and the result is shown in Fig. 5. The correlation coefficient between the two sets of concentrations is 0.76 (p < 0.05). Average concentrations for each PAH congener among all 13 street dust samples were calculated and compared with those among all 13 surface soil samples (Fig. 5), and shows a strongly positive correlation (R = 0.96, p < 0.001). This suggests that PAHs in surface soil and street dust have the same or similar sources in urban areas. Figure 6 shows that the top three PAH congeners found in all dust and soil samples were Flu, Pyr, and Phe, which account for 38% for dust samples and 34% for soil samples of all PAH concentrations, respectively. This result was consistent with a previous report that these

Table 1Concentrations of $\Sigma$ PAHs (total, 25 PAHs) instreet dust and surface soil fromdifferent areas of Dalian

Sampling area	Sample no.	$\Sigma$ PAHs (ng/g) in street dust		$\Sigma$ PAHs (ng/g) in surface soil	
		Mean	Range	Mean	Range
Garden	2	$3,540 \pm 2,327$	1,894–5,185	4,391 ± 4,370	1,301–7,481
Business/residence	7	$5,830 \pm 2,583$	1,970–10,117	$3,237 \pm 2,735$	649–8,541
Industry	4	$12,266 \pm 5,434$	5,071-17,074	$13,\!068 \pm 12,\!521$	2,751-28,904
All	13	$7,\!458 \pm 4,\!794$	1,894–17,074	$6,439 \pm 8,115$	649–28,904



Fig. 4 Relationship between total PAH concentration and TOC content in street dust and surface soil



Fig. 5 Total PAH levels in street dusts  $(C_D)$  versus surface soils  $(C_S)$ 



Fig. 6 Composition of PAH congeners in street dust and surface soil samples

three compounds accounted for about 40% of the total PAH content in three European cities (Morillo et al. 2007). Figure 6 also shows that PAHs in street dust and surface soil were dominated by higher molecular weight PAH (HMW-PAH) homologues (with four or more four rings). HMW-PAH accounted for about 73% and 72% of  $\Sigma$ PAHs in street dust and surface soil samples, respectively. Low

molecular weight PAHs (LMW-PAHs) were the minority, which was dominated by Nap, 1-MN, 2-MN, and Acy. This was relevant to their source and physicochemical characteristics. These LMW-PAHs mainly came from oil leaks and were easier to volatilize and degrade (Wang et al. 1999a). Nap, 1-MN, and 2-MN contributed more to the LMW-PAHs in street dust and surface soil because these three congeners have higher percentages than other LMW congeners in oil.

Statistical paired *t*-test was performed to analyze the similarity between compositions of PAH congeners in street dust and surface soil samples. The null hypothesis was that there is no difference between  $C_D$  and  $C_S$ , i.e.,  $H_0$ :  $C_D = C_S$ . K-S one-sample test verified whether a population was normally distributed. It was found both  $C_D$  and  $C_S$  are normally distributed. The *t*-test statistic was calculated to be t = 0.63. The sample number n = 13, thus at a 0.05 level of significance (*p*), the critical *t*-test value is 2.18. As  $*t^* < 2.18$ , the null hypothesis,  $H_0$ , cannot be rejected at p = 0.05. That is, there is no strong evidence to prove that  $C_D$  differs from  $C_S$ . Thus total PAHs exhibit similar concentrations in street dust and surface soil.

#### Sources of PAHs in Street Dust and Surface Soil

To assess and reduce the environmental risk caused by PAHs through source control, quantitative understanding the sources of PAHs is necessary. PAHs have natural and anthropogenic sources, and anthropogenic sources were the main ones in urban areas (Wilcke 2007). Anthropogenic sources of PAHs can be categorized into pyrogenic and petrogenic groups (Wang et al. 1999a; Yunker and Macdonald 2003). With pyrogenic sources, PAHs are generated by combustion of fossil fuel (coal and petroleum) and biomass. Another important primary source of PAHs contained in crude oil and petroleum products is petrogenic.

Diagnostic ratio is one widely used technique to apportion the origins of PAHs presented in different environmental media (Yunker et al. 2002; Bucheli et al. 2004). For example, BaA/(BaA + Chr) ratio values of  $\leq 0.35$  (diesel oil or kerosene = 0.35, crude oil = 0.12) suggest a petrogenic origin, those >0.40 (wood or grasses = 0.46, gasoline = 0.36, diesel oil = 0.38, brown coal = 0.44) indicate a pyrogenic origin, and results between these two critical values imply mixed origins. The ratio of Ant/(Ant + Phe) can also be used to identify petroleum- or combustion-origin PAHs. Ratio values < 0.10 (diesel oil = 0.09, kerosene = 0.04, crude oil = 0.07) are indicative of a petroleum origin, and those >0.10 (wood = 0.19, grasses = 0.17, gasoline or diesel oil = 0.11) are indicative of a pyrogenic origin (Yunker et al. 2002).

Figure 7 presents the ratios of BaA/(BaA + Chr) and Ant/(Ant + Phe) in street dust and surface soil samples



Fig. 7 Compositional relationship between BaA/(BaA + Chr) and Ant/(Ant + Phe) in street dust and surface soil from Dalian

from Dalian. The values of the BaA/(BaA + Chr) ratio ranged between 0.40 and 0.55 in soil, and from 0.35 to 0.50 in dust, suggesting that PAHs in soil were of pyrogenic origin, and the major portion of PAHs in dust was derived largely from pyrogenic sources such as soil. There were small portion of PAHs from Sites B/R4, I3, and I4 which had both pyrogenic and petrogenic origins. It is interesting to note that Sites I3 and I4 are petroleum industrial areas. Figure 7 also shows that ratios of Ant/(Ant + Phe) in all soil samples were >0.15, indicating that pyrogenic combustion may be the major origin of PAHs in soil. The ratios of all dust samples except the one from Site I4 were >0.10, indicating a pyrogenic origin. It is not surprising that PAHs in I4 had a petroleum source as indicated by the two ratios since I4 is a petroleum company.

In general, the major sources in street dust and surface soil are similar; PAHs are dominated by pyrogenic origins. The source difference between dust and soil is that petrogenic sources exist in street dust but not in soil. The reason for this difference is well understood and may be related to differences in location characteristics of dust and soil. Street dust is always located at the surface of the street and has more chance than surface soil to contact unburned or leaked fuel from automobiles in the street. It can therefore be concluded that the location difference between street dust and surface soil induces the different sources of PAHs in the two environmental media.

Principal component analysis (PCA) has been used to apportion the sources of PAHs in street dust and surface soil. PCA is a multivariate statistical technique that can be used in complex residue analysis to reveal relationships and patterns within datasets, which has been widely applied to discriminate the different origins of environmental pollutants (Zuo et al. 2007). Three principal components (PC1, PC2, and PC3) were extracted, with eigenvalues >1, and PC1, PC2, and PC3 explained 64.4%, 22.%, and 5.6% (92.9% in total) of the total variance in dust and for 85.8%, 6.0%, and 4.5% (96.3% in total) of the total variance in soil.

Figure 8 presents the results of PC1 and PC2 for PAHs in street dust and surface soil samples, and as shown, PC1 and PC2 can explain most of the variation in dust (87.3%) and soil (91.8%). It can be seen in Fig. 8 that HMW-PAHs (4-6 rings) constituted PC1 in street dust, and PC2 was dominated by LMW-PAHs (2-3 rings). HMW-PAHs are caused by pyrogenic sources (fossil fuel [coal and petroleum] combustion), because of a strongly positive correlation between HMW-PAHs and high-temperature combustion (Wang et al. 1999a, b; Soclo et al. 2000). On the contrary, LMW-PAHs (2-3 rings) can be attributed to petrogenic sources, which come from volatilization and leak of crude oil and petroleum products (Wang et al. 1999a, b; Soclo et al. 2000). A plot of PC1 and PC3 for street dust (not shown) suggests that PC3 is mostly associated with Ret and Thn, Ret being a molecular marker of biomass combustion (Ramdahl 1983; Simcik et al. 1999; Larsen and Baker 2003). Thus, in street dust, PC1, accounting for 64.4% of the total variance, can be considered representative of pyrogenic sources; PC2 (29.9%), of petrogenic sources; and PC3 (5.6%), of biomass

Fig. 8 Diagrams of factor loadings of first two principal components (PC1 and PC2) for PAHs in (a) street dust and (b) surface soil



combustion sources (which also are pyrogenic sources). That is, pyrogenic sources collectively accounted for 70.0% of the total PAHs in street dust. These results are consistent with a previous study showing that the PAH contribution percentages of pyrogenic and petrogenic sources in road dust of Shanghai were 71.0% and 11.4% in winter, and 64.9% and 14.1 in summer, respectively (Liu et al. 2007).

PAH profiles in surface soil were quite different. PC1 accounts for larger than loading of PC1 in street dust. Figure 8 shows that PC1, with 85.8% of the total variance, is highly loaded by all the PAHs except Ret and Pyr. This probably implies that the sources of PAHs in soil are pyrogenic because PAHs are dominated mainly by HMW-PAHs. PC2 is mainly associated with Pyr, which might be related to gasoline engine emission (Christensen and Bzdusek 2005). Similar to PAHs in dust, PC3 is also associated with Ret, and biomass combustion accounts for 4.5% of the total variance in soil. These results indicate that PAHs in soil all come from pyrogenic sources and no petrogenic sources are found in soil of Dalian, which is consistent with a previous study (Wang et al. 2007).

Although urban PAH concentrations can be influenced by local and long-distance transport sources, local contributors, such as industrials and automobiles, are the major sources (Zheng and Fang 2000; Hou et al. 2006; Tamamura et al. 2007). To estimate the distance of potential sources, the ratio of BaA/Chr was adopted as a diagnostic indicator. The ratio of BaA/Chr was used to evaluate the origin of PAHs. A lower BaA/Chr ratio implies a longer distance of transport, while a higher ratio suggests that those PAHs might be mostly from emission of local sources (Mai et al. 2003). Since BaA is photodegraded and oxidize degraded more easily than its isomer Chr during their atmospheric travel (Kamens et al. 1988; Fraser et al. 1998; Esteve et al. 2004), the BaA/Chr ratio can be used as a tracer for the degree of photodegradation and oxidized degradation, in addition to being used as a source discriminator. If the ratio is low, the sources may be far away from the urban areas because of the photodegradation and oxidized degradation of most BaA, while a high ratio is referred to a local source. Mai et al. (2003) found that the BaA/Chr ratios of Macao street dust were in the range of 0.2-0.3 and suggested that Macao street dust might be transported from distant areas. On the contrary, Liu et al. (2007) reported that the BaA/Chr ratio range of Shanghai road dust was 0.6–1.4, which meant that Shanghai might be close to its potential sources of dusts. BaA/Chr values are in the range of 0.6–0.9 in street dust and 0.7–1.0 in surface soil of Dalian, and this range is close to the range of Shanghai road dusts, which suggests that PAHs in street dust and surface soil in Dalian come mostly from emission of local sources.

#### Conclusion

Concentrations of PAHs in street dust and surface soil in the city of Dalian in 2007 have been studied, and the results show that PAHs exhibit similar concentrations in street dust and surface soil. Mean PAH concentrations in both dust and soil were much higher at industrial sites than at business/residential or garden sites. PAHs were dominated by higher molecular weight PAH (4- to 6-ring) homologues, which accounted for about 73% and 72% of total PAHs in street dust and surface soil, respectively. No apparent correlations were found between the concentrations of PAHs in street dust and TOC contents, whereas the concentrations of PAHs in surface soil showed a strong correlation with the TOC content of soil. Pyrogenic and petrogenic sources contributed 70% and 22.4% of the total PAHs in street dust, in which fossil fuel (coal and petroleum) and biomass combustion accounted for 64.4% and 5.6% of the total PAHs from pyrogenic sources, respectively. In surface soil, the total PAHs were dominated by pyrogenic sources. PAHs in dust and soil may come mainly from local sources on the basis of the diagnostic ratios of BaA/Chr.

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