

# Distribution of Polycyclic Aromatic Hydrocarbons in Soils of an Arid Urban Ecosystem

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Received: 11 June 2010 / Accepted: 8 December 2010 / Published online: 30 December 2010  
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**Abstract** Polycyclic aromatic hydrocarbons (PAHs) are widespread environmental pollutants produced by incomplete combustion sources such as home heating, biomass burning, and vehicle emissions. PAH concentrations in soils are influenced by source inputs and environmental factors that control loss processes and soil retention. Many studies have found higher concentrations of these pollutants in soils within cities of temperate climates that have a centralized urban core. Less is known about the factors regulating PAH abundance in warm, arid urban ecosystems with low population densities but high traffic volumes. The relative importance of sources such as motor vehicle traffic load and aridland ecosystem characteristics, including temperature, silt, and soil organic matter (SOM) were explored as factors regulating PAH concentrations in soils near highways across the metropolitan area of Phoenix, AZ (USA). Highway traffic is high compared with other cities, with an

average of 155,000 vehicles/day. Soils contained low but variable amounts of SOM (median  $2.8 \pm 1.8\%$  standard deviation). Across the city, median PAH concentrations in soil were low relative to other cities,  $523 \pm 1,886 \mu\text{g/kg}$ , ranging from 67 to 10,117  $\mu\text{g/kg}$ . Diagnostic ratio analyses confirmed that the source of PAHs is predominantly fuel combustion (i.e., vehicle emissions) rather than petrogenic, biogenic, or other combustion sources (coal, wood burning). However, in a multiple regression analysis including traffic characteristics and soil properties, SOM content was the variable most strongly related to PAH concentrations. Our research suggests that dryland soil characteristics play an important role in the retention of PAH compounds in soils of arid cities.

**Keywords** Sonoran desert · Soil organic matter · Urban ecosystem · PAH · Carbon deposition · Arid

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## 1 Introduction

Human activity produces chemicals that deteriorate environmental quality and can be harmful to organisms (US EPA 1998). The negative ecological consequences and increased public awareness about the impacts of point-source contamination have stimulated legal control at sites with concentrated toxins. However, the rise in motor vehicle usage has raised concerns about non-point, diffuse pollution that occurs at lower intensities but covers larger

areas (Johnsen and Karlson 2007). The United States Environmental Protection Agency (EPA) and the United Nations Environment Programme (UNEP) placed various combustion-derived compounds associated with anthropogenic sources under regulation in the 1990 US Clean Air Act and in the 1995 Stockholm Convention on Persistent Organic Pollutants (US EPA 1998; 2005). Polycyclic aromatic hydrocarbons (PAHs), also known as polynuclear aromatic hydrocarbons, are some of the most widespread pollutants in the environment, identified in both the US EPA Priority Pollutants and UNEP lists. These chemicals are by-products of incomplete combustion processes from common sources including wood burning, food cooking, vehicle engine emissions, industrial exhaust, and cigarette smoke (Rogge et al. 1993; Cass 1998; Srogi 2007; Nam et al. 2009). The effects of PAH compounds include human immunotoxic responses and ecological changes through biological and physico-chemical interactions (ATSDR 1995).

Based on studies carried out in predominantly temperate climates, a diverse suite of factors affect the abundance and distribution of PAHs in the environment. Concentrations are generally highest near sources and decline with distance (Yang et al. 1991; Bryselbout et al. 2000; Tuhackova et al. 2001; Glaser et al. 2005; Olson and McDow 2009). Once deposited onto soils, PAHs can be degraded by biotic or abiotic processes (e.g., microbial, chemical), re-volatilized into the atmosphere, or transferred to soil profiles, groundwater, and aquatic systems through runoff and leaching (Juhász and Naidu 2000; Wilcke 2000; Lima et al. 2005; Nadal et al. 2006; Haritash and Kaushik 2009). The physico-chemical interactions between PAHs and the environment affect transformation, movement, and losses from the soil system. For example, temperature is positively related to PAH chemical reactivity (Dabrowska et al. 2005; Zhang et al. 2005). Additionally, these compounds may adsorb to and be stabilized by mineral surfaces and organic matter (Cornelissen et al. 2005). While the environmental fate of anthropogenic chemicals is diverse, studies have shown that soil PAH concentrations are higher in urbanized and industrial use locations compared with rural areas (Wilcke 2007). Contamination appears to be localized primarily near sources, for instance, near dense, highway corridors in cities with a centralized urban core (Van Metre et al. 2000;

Wilcke 2000). However, these results may also reflect the abundance of studies that explore the fate of PAH compounds in polluted sites within older cities in cool temperate climates. By comparison, less is known about the fate of PAH compounds in warm arid and semi-arid ecosystems with urban-sprawl development, despite high traffic loads and rapid urban growth expected for these regions (Warren et al. 1996; Jenerette and Wu 2001, FHA 2008).

In addition to the importance of source inputs, other features such as soil properties and environmental conditions that characterize dryland ecosystems may affect PAH content in soils of warm, arid cities. For example, soils in arid climates receive intense solar radiation, which can photolyse PAH compounds to intermediate quinones, lower-mass PAH compounds, or degrade them completely (Guieysse et al. 2004; Dabrowska et al. 2005; Nadal et al. 2006). This photodegradative process may transform recalcitrant compounds into other forms that are more available for microbial degradation (Guieysse et al. 2004). Additionally, temperatures can reach  $>65^{\circ}\text{C}$  on mineral surfaces during the summers (McCalley and Sparks 2009), facilitating volatilization of low-molecular weight PAH compounds back to the atmosphere (Park et al. 1990; Paasivirta et al. 1999). Arid deserts contain soil organic matter (SOM) in low concentration (Zhu et al. 2006) and receive high loads of dust, which is composed of fine-textured particles that are predominant in the silt fraction (Pewe et al. 1981; Simonson 1995; Muhs et al. 2008; Lawrence and Neff 2009). Furthermore, low SOM content may lead to low rates of PAH retention, as these pollutants readily adsorb to organic surfaces (Bucheli et al. 2004; Cornelissen et al. 2005). Low-organic matter content of dryland soils leads to carbon (C) limitation of heterotrophic soil microorganisms that may metabolize organic anthropogenic pollution as an energy source (Kaye et al. *in revision*; Semple et al. 2007; Peng et al. 2008). As a result, the microbial community structure may be modified, reflecting a dominance of urban-adapted microbes that may decrease PAH loads in soil (Leys et al. 2005; Johnsen and Karlson 2005; Van der Meer 2006). PAH compounds are susceptible to biodegradation on silt-sized particles (Hwang and Cutright 2004), but high silt content in desert soils may also increase pollutant retention because PAHs preferentially adsorb to aromatic structures within the silt fraction (Wilcke

2000; Muller et al. 2001; Krauss and Wilcke 2002; Ni et al. 2008).

Despite the multitude of studies on PAH occurrence and fate in the environment, there remains a gap in the understanding of the relative importance of sources and environmental factors that control the fate of PAH compounds in soils within rapidly expanding arid cities. In the present work, PAH concentrations were determined in surface soils along major highways in the Phoenix, AZ metropolitan area. Concentration patterns are discussed relative to vehicle traffic load, road characteristics, and a suite of soil properties.

## 2 Materials and Methods

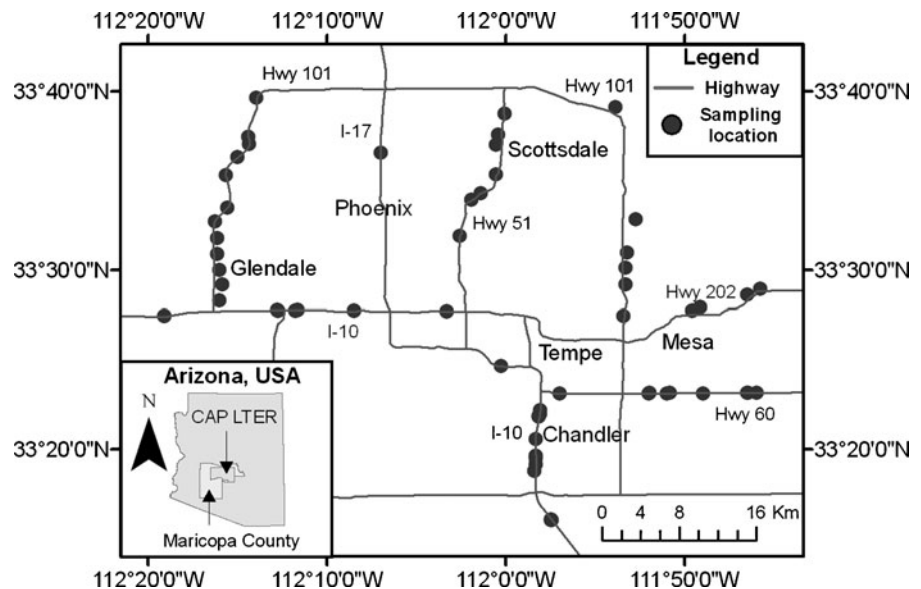
### 2.1 Study Area Description

Surface soil samples were collected along highway corridors in the 2,400-km<sup>2</sup> Phoenix metropolitan area in Arizona (Fig. 1), within the boundaries of the 6,400-km<sup>2</sup> Central Arizona–Phoenix Long-Term Ecological Research project (CAP LTER; <http://caplter.asu.edu>). The Phoenix metro area is located in the Sonoran Desert. Climate within the study region is characterized by hot summers (average 34°C) and mild winters (average 14°C), with a mean annual temperature (MAT) of 23°C (1971–2000; NOAA 2008). Surface temperatures in the summer range

from 26°C at night to 52°C during the day (Buyantuyev and Wu 2010), with an average of 37°C (Green and Oleksyszyn 2002). Because of its low latitude and the prevalence of clear, dry skies, Phoenix soils are exposed to high levels of visible and UV irradiance, receiving on average 85% of the annual total possible sunshine (Cervany 1996). Sonoran Desert rainfall is distributed mainly with summer monsoon events from July to September and storms from the Pacific Ocean between November and March (WRCC 2009). Mean annual precipitation is 193 mm but is highly variable from year to year (NOAA 2009). Dust storms often occur during the summer and are associated with thunderstorm activity (Nickling and Brazel 1984), dropping on average 54.5 gm<sup>-2</sup> year<sup>-1</sup> of dust, 75% of which contains silt-sized particles (Pewe et al. 1981; Simonson 1995; Lawrence and Neff 2009).

Low-density development and high vehicle traffic loads are common in cities similar to those in the southwestern USA, which are characterized by a unique suite of anthropogenic and environmental factors that may influence the fate of organic pollutants. The Phoenix metropolitan area is characterized by relatively new highways with an average age of 24.5 years. The maximum traffic volume reaches up to 300,000 vehicles/day (both directions) on the Interstate-10 highway in Phoenix, which is comparable to some of the most traveled roadways in all of Europe (United Nations 2003). Additionally, this Interstate-10 highway segment ranks 21st highest

**Fig. 1** Soil sampling locations ( $N=60$ ) near highways in the Phoenix metropolitan area. The study area is within the CAP LTER region, located in Maricopa County, AZ. Site markers overlap where sampling areas were at the same highway segment but on opposite sides of the roadway



out of all major USA highways (~300 total) in average daily traffic, with ten highways out of the top 25 occurring in Los Angeles, a similarly low-density metro area (FHA 2008). The average total traffic load is 155,400 vehicles/day, composed of 131,500 automobiles and 23,900 trucks. The Phoenix metro area contains 4.3 million people, with a density of 660 people/km<sup>2</sup> (US Census 2008). Urban land cover has expanded by 54 km<sup>2</sup>/year (1970–2000; Keys et al. 2007), and human population has increased exponentially over the past 100 years (Jenerette and Wu 2001, US Census 2005; 4.5% between 1990 and 2000), similar to rates of growth in other arid and semi-arid cities globally (Warren et al. 1996; Alshuwaikhat and Nkwenti 2002). The highway system in the metro area includes seven distinctive segments covering multiple municipalities. Each segment varies considerably in roadside landscaping and soil characteristics, such as presence of surface gravel, visible calcium carbonate from exposed subsurface soil horizons, aspect, slope of soil away from the road, vegetation composition and density, irrigation, and roadside curb barriers.

## 2.2 Soil Sample Collection

Prior to the monsoon rains in July 2008, 60 sites were chosen randomly across the Phoenix metro area along major highways that ranged widely in age and traffic volume. To isolate the effect of highway traffic as a main PAH source, sampling was avoided along highway segments that occurred within 20 m of other roads, parking lots, industrial buildings, residential areas, or other potential PAH sources. Sampling was avoided along highway segments that were surrounded by pavement (no soil) or were under construction. At each site, three soil cores (each separated by 0.5 m) were collected to 2 cm depth at a distance of 0.5 m away from the road. Care was taken to sample >5 m away from shrubs. Replicate cores per site were homogenized in a polyethylene freezer storage bag for analysis of soil properties. Samples were collected and immediately stored into a 250-mL tight-sealed amber glass jar (baked overnight at 450°C prior to use) for PAH analysis. After sample collection, all soils were placed in a cooler with ice packs to minimize post-collection microbial transformations of soil compounds. Samples used for soil properties were dry upon collection and further air

dried and stored for up to 24 h before sieving (to 2 mm) and for up to 5 days before being processed. Soil samples collected for PAH analysis were not air dried, but were promptly sieved to 2 mm in the laboratory and stored in amber jars at -16°C freezer temperature until PAH sample preparation.

In addition to investigating the distribution of PAH compounds across the city, PAH abundance was explored at smaller spatial scales near roadways. At 5 of the 60 random sites, three 15 m transects were identified perpendicular to the road, each separated by 0.5 m. Along each transect, one soil sample (0–2 cm depth) was collected at 0.5, 1.5, 5, and 15 m. Samples at each distance within each site were composited and homogenized ( $N=5$  replicate sites  $\times$  4 distances per site). Samples were processed similar to other soils and analyzed for PAH compounds and soil properties according to methods below.

## 2.3 Soil Properties and Roadway Characteristics

A suite of physical and chemical properties of soils were measured on each soil sample, as these are known to influence the fate and retention of organic compounds. Soil laboratory methods were based on standard protocols used in the CAP LTER network (McCrackin et al. 2008; Hall et al. 2009). Gravimetric soil moisture (%) was determined by drying 30 g of soil for 24 h in a 105°C oven. SOM (g organic matter per 100 g of dry soil; %) was determined by the loss-on-ignition method (Storer 1984) as ash-free dry mass following combustion of oven-dried soils for 4 h at 550°C. Particle size was determined using the hydrometer method (100 mL of 50 gL<sup>-1</sup> sodium hexametaphosphate in 40 g of soil), followed by sieving (to 53  $\mu$ m) for sand content and calculating silt content by difference. Prior to soil texture analysis, carbonate was removed using 10 mL of 1 M sodium acetate (pH 5) in 100 mL of DI water. Soil water holding capacity (WHC; %) was measured by saturating 20 g of soil with water and weighing after a 24-h drain time through a GF-A filter. Ammonium ( $\mu$ g NH<sub>4</sub><sup>+</sup>-N  $\cdot$  g<sup>-1</sup> dry soil) and nitrate + nitrite (summed as  $\mu$ g NO<sub>3</sub><sup>-</sup>-N  $\cdot$  g<sup>-1</sup> dry soil) concentrations were measured using 10 g of soil that was extracted in 50 mL of 2 M KCl by shaking for 1 h and filtered through pre-leached Whatman #42 ashless filters. The extracts were frozen until colorimetric analysis using a Lachat Quickem 8000

autoanalyzer. Potential rates of two soil microbial processes (net N mineralization and net nitrification) were assessed by incubating 10 g of soil in the dark at 20°C for 10 days at 60% WHC, followed by extraction with 2 M KCl and colorimetric analysis for inorganic N as described above. Rates of potential net N mineralization and net nitrification were calculated as the difference in the sum of  $\text{NH}_4^+$  and  $\text{NO}_3^-$ , or  $\text{NO}_3^-$  alone, respectively, before and after incubation divided by the number of incubation days (reported as  $\mu\text{g N} \cdot \text{g}^{-1} \text{day}^{-1}$ ).

Traffic load and highway age data were gathered from databases provided by the Arizona Department of Transportation and Maricopa Association of Governments (MAG 2007, ADOT 2008). Traffic load (automobiles, trucks, and total) was calculated from annual data as the daily number of vehicles driving both directions in all lanes of the road segment. Highway age is the number of years since the highway was constructed or since a road was upgraded to a highway.

## 2.4 PAH Content Determination

PAH concentrations were determined following common methodologies (e.g., Wilcke 2007, US EPA 1996, 2007). After soil collection and freezer storage, 20 g of soil from each site was extracted three times with 20 ml dichloromethane (Optima Grade DCM; Fisher Scientific, Fairlawn, NJ) under sonication for 20 min. The combined extracts were filtered through a QM-A grade quartz microfiber filter using a glass syringe and cleaned over a silica column (Supelco Omnifit 250×10 mm). The silica used was Merck grade 7754 (70–230 mesh, 60 Å; Sigma-Aldrich, St. Louis, MO), pre-conditioned with DCM. The samples were eluted using DCM. The eluate was concentrated down to 250  $\mu\text{L}$  under a gentle stream of high purity nitrogen and stored in a freezer (−16°C) until analysis.

Samples were analyzed using a Shimadzu gas chromatography–mass spectrometry system (GC-17a, GC/MS-qp5000) equipped with a DB-5 MS (Agilent) fused-silica capillary column. A 1  $\mu\text{L}$  aliquot was injected in splitless mode, and elution was performed using helium carrier gas at a flow rate of 1.2 mL/min. The injection and interface temperature were set at 300°C and 275°C, respectively. The initial temperature was set at 60°C, holding for 10 min, then increased to 300°C at 10°C/min, with the final

temperature being held for 20 min for a total run time of 54 min. MS detection was set in Scan acquisition mode with a start and end mass-to-charge ( $m/z$ ) ratio of 40 to 350, respectively. Deuterated PAH internal standards (naphthalene-d8, pyrene-d10, chrysene-d12, and perylene-d12; Sigma-Aldrich, St. Louis, MO) were spiked into the samples prior to the extraction procedure to quantify PAHs and to account for losses during sample preparation and processing. A PAH Semi-volatiles Calibration CLP Mix (Supelco, Bellefonte, PA) was used as a calibration standard. PAH compound identification and peak integration were performed manually for all samples. Quality control procedures included calibration checks and the use of laboratory blanks throughout the entire preparation procedure.

Twenty compounds were quantified, including 16 PAHs from the US EPA Priority Pollutants list (referred to as  $\Sigma 16\text{PAHs}$  here). Seven of the listed PAH compounds marked below with an asterisk (\*) are classified by EPA as probable human carcinogens (referred to as  $\Sigma 7\text{PAHs}$  here). The 16 compounds measured were: naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FTH), pyrene (PYR), \* benzo(a)anthracene (BaA), \* chrysene (CHR), \* benzo(b)fluoranthene + \* benzo(k)fluoranthene (BbkF), \* benzo(a)pyrene, \* indeno(1,2,3-cd)pyrene (IcdP), \* dibenzo(a,h)anthracene (DahA), and benzo(g,h,i)perylene (BghiP). The two PAHs, benzo(b)fluoranthene and benzo(k)fluoranthene, were summed together because they presented substantial overlap in the chromatograms. In addition to the  $\Sigma 16\text{PAHs}$ , four PAH compounds were determined, including acephenanthrylene (ACP), benzo(j)fluoranthene (BjF), benzo(e)pyrene (BeP), and perylene (PER). Finally, seven other PAH compounds were quantified, which are methyl homologues of naphthalene, phenanthrene, and anthracene (MPheAnt). Raw data of individual PAH concentrations, soil characteristics, and road site properties for all study sites are available on the CAP LTER database (<http://caplter.asu.edu/data>).

## 2.5 Diagnostic Ratios of PAH Compounds for Source Identification

Diagnostic ratios (DRs) using relative concentrations of individual PAH compounds (i.e., NAP, ACY, etc.), grouped PAHs (i.e., low-molecular weight [LMW; molecular mass <202], high-molecular weight



[HMW; molecular mass  $\geq 202$ ]), and specific marker compounds can reveal their source (Yunker et al. 2002). These indicator ratios have been determined from controlled lab experiments or from field assessments based on known sources and historic changes (Rogge et al. 1993; Yunker et al. 2002; Lima et al. 2005). The DRs calculated in this study can be used to distinguish between various anthropogenic machinery and natural sources, including combustion from diesel, gasoline, or wood burning. To identify sources, the following commonly used DRs were calculated: (a) IcdP/(IcdP + BghiP):  $<0.2$  = petroleum,  $0.2-0.5$  = gasoline combustion, and  $>0.5$  = other pyrogenic sources such as wood combustion (Yunker et al. 2002); (b) ANT/(ANT + PHE):  $<0.1$  = petrogenic and  $>0.1$  = pyrogenic (Wang et al. 2009); (c) BaA/(BaA + CHR):  $<0.2$  = petrogenic and  $>0.35$  = combustion (Yunker et al. 2002); (d) FTH/(FTH + PYR):  $<0.4$  = petrogenic and  $>0.5$  = non-gasoline combustion (e.g., diesel or coal),  $0.4-0.5$  = fuel combustion (Yan et al. 2006); and (e) (MPheAnt)/PHE:  $2-6$  = petrogenic,  $<1$  = gasoline, and  $1-2$  = diesel (Hwang et al. 2003; Bucheli et al. 2004). The ratio of IcdP/(IcdP + BghiP) is the most reliable to be used for source diagnostics due to its inclusion of HMW PAHs that undergo significantly less modification after production compared to the LMW PAHs that re-volatilize from the soil (Park et al. 1990; Kaupp and McLachlan 1999; Bucheli et al. 2004; Zhang et al. 2005; Brandli et al. 2008). The other DRs are less reliable in a warm environment, but are included here because they discriminate between sources of combustion, such as vehicle fuel from others such as coal or wood burning, and between pyrogenic (combustion) and petrogenic (non-combusted crude oil) sources. Additionally, two sets of ratios, BaA/(BaA + CHR) and IcdP/(IcdP + BghiP), were utilized in a cross-plot to further identify sources. Finally, “signature” PAH markers were identified within the soils, the dominance of which indicates production from specific motor vehicle sources, including BghiP (non-catalyst automobile engine emissions) and PHE (heavy-duty diesel truck emissions; Rogge et al. 1993; Ravindra et al. 2008).

## 2.6 Data Analysis

SPSS 17 software was used for all statistical analyses. Raw data were tested to meet assumptions for

parametric statistical analysis. All data except traffic load and soil pH were  $\log_{10}$ -transformed (median values reported for descriptives) to achieve normality, homoscedasticity, and linearity. Adjusted coefficient of determination ( $r^2$ ) was used to evaluate the strength of independent variables controlling PAH concentrations. Non-correlated variables were entered into a backward stepwise multiple linear regression to evaluate effects of soil properties and traffic characteristics on PAH concentrations in soils. Similarly, regression analysis was used to evaluate the effects of soil properties and distance away from the road. PAH concentrations were divided by SOM concentrations to test for the effects of distance but eliminating the influence of SOM. PAH/SOM values (reported as  $10^{-8}$  g PAH  $\cdot$  g $^{-1}$  SOM) were non-normal with unequal variance and were square root-transformed in order to meet assumptions for parametric analysis. A one-sample Student's *t* test was used to evaluate the significance of PAH concentrations in roadside soil of Phoenix compared with other cities.

## 3 Results

### 3.1 Highway Soil and Road Characteristics of the Phoenix Metro Area

Soil characteristics were highly variable across the heterogeneous urban highway environment. SOM ranged from 0.8% to 9.9%, with a median of 2.8% ( $\pm 1.8\%$  standard deviation). Soils were generally alkaline, ranging from pH 7.2 to 9.5 with a mean pH of  $8.2 \pm 0.5$ . Particle size of soils was on average 68.5% sand, 24.5% silt, and 7% clay, with a median WHC of  $24.8 \pm 7.3\%$ . Median  $\text{NH}_4^+$  and  $\text{NO}_3^-$  concentrations were  $6.1 \pm 23.5$  and  $18.8 \pm 88.5$   $\mu\text{g N} \cdot \text{g}^{-1}$ , respectively. Potential net N transformations were highly variable: median potential net N mineralization was  $0.3 \pm 11.4$   $\mu\text{g N} \cdot \text{g}^{-1} \text{day}^{-1}$  and median potential net nitrification was  $0.8 \pm 10.9$   $\mu\text{g N} \cdot \text{g}^{-1} \text{day}^{-1}$ . Soil moisture was low due to sampling during the hottest summer month of the year, with a median of  $0.4\% \pm 0.5\%$ . These soil properties and processes are in the range of those measured in prior ecological studies in the CAP LTER (Zhu et al. 2006; McCrackin et al. 2008; Hall et al. 2009).

### 3.2 Soil PAH Concentrations and Sources

The total PAH concentration in soils, particularly LMW PAHs, was expected to be low relative to other cities due to the high summer temperatures of the Phoenix environment. Mean  $\Sigma 16\text{PAHs}$  was  $1,174 \pm 1,886 \mu\text{g} \cdot \text{kg}^{-1}$  SD, ranging from 67 to  $10,117 \mu\text{g} \cdot \text{kg}^{-1}$ . The median PAH concentration was  $523 \mu\text{g} \cdot \text{kg}^{-1}$  (Table 1), reflecting a positively skewed distribution. HMW compounds dominated the PAH profile (91% of total). In an analysis of all published studies on PAH concentrations in roadside

soils, average PAH concentration in Phoenix is lower than in other cities ( $df=20$ ,  $t=2.58$ ,  $p=0.018$ ; Table 2), despite the fact that this metropolitan area experiences some of the highest traffic loads reported worldwide.

Diagnostic ratios and marker compounds indicate that PAHs in highway roadside soils of this arid city originate from anthropogenic sources, particularly diffuse, non-point emissions from gasoline and diesel engines. Ratios of  $\text{IcdP}/(\text{IcdP} + \text{BghiP})$ ,  $\text{BaA}/(\text{BaA} + \text{CHR})$ ,  $\text{FTH}/(\text{FTH} + \text{PYR})$ ,  $\text{ANT}/(\text{ANT} + \text{PHE})$ , and  $(\text{MPheAnt})/\text{PHE}$  all indicate that >90% of PAH

**Table 1** Median concentration ( $\mu\text{g}\cdot\text{kg}^{-1}$ ), relative contributions, and descriptives of PAHs in Phoenix highway soils

PAHs	Mass (MW)	No. of rings	Median concentration ( $\pm 1$ SD)	Min	Max	Percent of total
NAP	128	2	5 (9)	<1	40	1
ACY	152	3	5 (38)	<1	168	2
ACE	154	3	1 (2)	<1	10	<1
FLU	166	3	1 (4)	<1	27	<1
PHE	178	3	24 (81)	3	519	4
ANT	178	3	8 (42)	<1	180	2
FTH	202	4	79 (290)	5	1,867	14
ACP	202	4	3 (14)	<1	77	1
PYR	202	4	79 (272)	5	1,659	14
BaA	228	4	34 (193)	2	1,068	8
CHR	228	4	89 (285)	5	1,476	16
BbkF	252	5	40 (134)	3	605	8
BjF	252	5	8 (29)	<1	163	2
BeP	252	5	52 (152)	3	754	9
BaP	252	5	40 (168)	1	839	9
PER	252	5	12 (48)	1	268	2
IcdP	276	6	34 (170)	1	1,049	7
DahA	278	5	14 (71)	<1	481	3
BghiP	276	6	76 (261)	6	1,766	12
2-Ring			5 (9)	<1	40	1
3-Ring			42 (151)	5	767	9
4-Ring			275 (1,021)	27	5,814	52
5-Ring			90 (355)	9	1,703	20
6-Ring			114 (429)	7	2,815	19
LMW	128–178		45 (154)	5	773	9
HMW	202–278		474 (1,743)	59	9,344	91
Toxic $\Sigma 7\text{PAHs}$			246 (972)	26	4,906	51
Median $\Sigma 16\text{PAHs}$			523 (1,886)	67	10,117	100

Percent of total is contribution of each PAH out of  $\Sigma 16\text{PAHs}$ . ACP, BjF, BeP, and PER are not included in  $\Sigma 16\text{PAHs}$ .  $N=60$

ACE acenaphthene, FLU fluorene, BaP benzo(a)pyrene, DahA dibenzo(a,h)anthracene, ACP acephenanthrylene, BjF benzo(j) fluoranthene, BeP benzo(e)pyrene, PER perylene. See [Materials and Methods](#) text for names of PAH abbreviations

**Table 2** Comparison of all published studies investigating PAH concentration in roadside soils

Average PAH concentration ( $\mu\text{g}\cdot\text{kg}^{-1}$ )	Roads or samples ( <i>N</i> )	City, Country	Human census ( $10^6$ ) <sup>a</sup>	Vehicle traffic load ( $10^3$ ) <sup>a</sup>	Soil organic matter (%) <sup>b</sup>	Annual precipitation (mm)	Mean annual temperature ( $^{\circ}\text{C}$ )	References
40,140	7	Peshawar, Pakistan	2.5			400	22.0	Khan et al. 2008
20,900	20	Detroit, USA	4.0			820	9.5	Wang et al. 2008
14,230	7	Ji'nan, China	5.9		4.2	675	14.2	Dai et al. 2008
12,980	80	Agra, India	1.3	100		660	25.0	Masih and Taneja 2006
8,700	3	Copenhagen, Denm.	0.5	36		525	8.0	Johnsen et al. 2006
7,189	24	New Orleans, USA	1.1			1500	20.0	Wang et al. 2008
6,280	17	Shanghai, China	19.0			1162	18.1	Jiang et al. 2009
4,130	5	Kurashiki City, Japan	0.5	30		1158	16.0	Oda et al. 2001
3,902	3	Bayreuth, Germany	0.1	50		688	8.3	Glaser et al. 2005
3,346	4	Brisbane, Australia	1.3	36		1090	20.0	Yang et al. 1991
3,095	3	Prague, Czech Rep.	1.2	90		565	8.8	Tuhackova et al. 2001
2,870	4	Beijing, China	17.0			576	11.8	Chu et al. 2003
2,627	9	Eastern France		70		800	10.0	Crepineau et al. 2003
1,587	3	Siedlce, Poland	0.1	8		600	7.0	Kluska 2003
1,287	3	Tehran, Iran	10.0			242	16.5	Samimi et al. 2009
1,174	60	Phoenix metro, USA	4.3	155	3.4	193	23.0	Current study
887	9	Kathmandu, Nepal	1.6			1400	18.0	Aichner et al. 2007
785	11	Seville, Spain	1.4		1.9	533	18.6	Morillo et al. 2008
676	30	Chiang-Mai, Thailand	0.2			1126	26.0	Amagai et al. 1999
370	16	Poland			1.9	600	8.0	Maliszewska-K. et al. 2008; 2009
107		Bangkok, Thailand	5.6			1320	28.0	Wilcke et al. 1999
90	5	Fruska Gora, Serbia	0.4	8		580	11.0	Stankovic et al. 2008

<sup>a</sup> Human population and traffic volume reported during year of study.

<sup>b</sup> Soil organic matter data are missing for many of the studies reported here

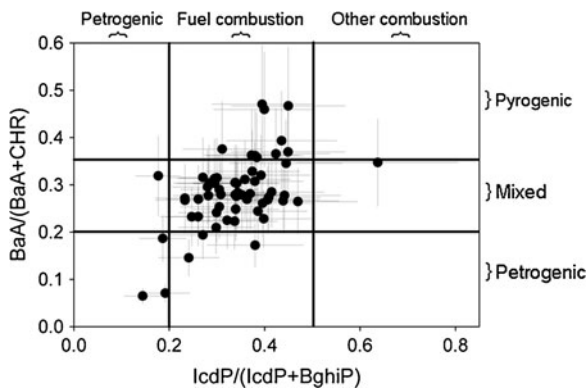
compounds in highway soils originate from motor vehicles. Furthermore, a cross-plot using a combination of IcdP/(IcdP + BghiP) and BaA/(BaA + CHR) confirms that PAHs predominately originate from fuel combustion in more than 80% of the sites sampled (Fig. 2). This conclusion is supported by marker compounds: on average, PHE contributed 4% total to the PAH profile, more than any other low-molecular weight PAH, while BghiP was also a significant contributor (12%) compared with other high-molecular weight PAHs (Table 1).

### 3.3 Relationship Between PAHs and other Variables

Soil properties associated with sorption play a significant role in the fate of PAHs after their

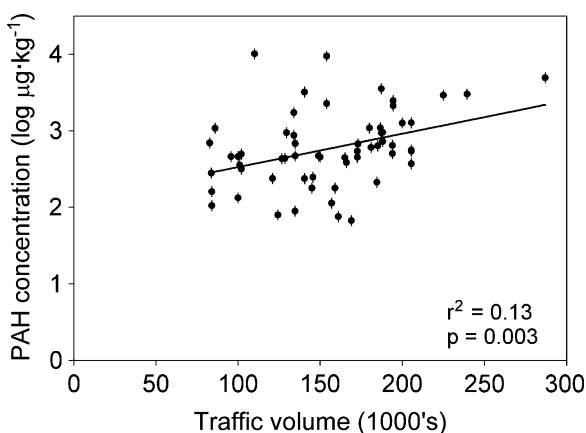
emission and deposition in the Phoenix environment. In bivariate regressions between PAHs and individual soil and traffic properties, PAH concentrations ( $\log_{10}$  transformed) were significantly but weakly related to vehicle traffic load (Fig. 3;  $r^2=0.13$ ,  $p=0.003$ ) and highway age ( $r^2=0.10$ ,  $p=0.007$ ). However, these relationships were weaker than the relationship between PAHs and SOM (Fig. 4;  $r^2=0.39$ ,  $p<0.001$ ). In a multiple regression analysis using all road characteristics and soil properties as independent variables and  $\log_{10}$  PAH concentrations as the dependent variable, both SOM and traffic load were the only variables significantly related to PAH concentrations in soil ( $r^2=0.47$ ,  $p<0.001$ ), with SOM as the more important of the two (Table 3;  $\beta$  coefficients=0.55 and 0.31, respectively). Silt content



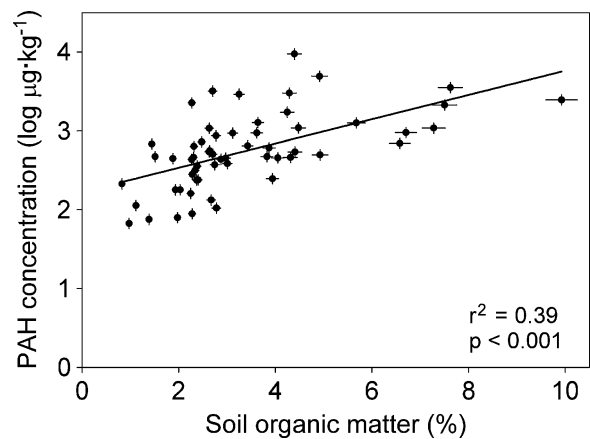


**Fig. 2** Diagnostic ratios IcdP/(IcdP + BghiP) and BaA/(BaA + CHR) used in a cross-plot, showing a dominance of vehicle emission sources. The grid lines represent source identification based on literature values. Other combustion includes wood, coal, and non-fuel burning processes. Mixed includes diesel engine, automobile emissions, and combusted engine oil sources. Each point represents one sample consisting of three homogenized soil cores, showing bidirectional error bars for analytical error

was not significantly related to PAH concentrations. In an analysis of distance-from-highway data using all soil and roadway characteristics, SOM was the only variable significantly related to PAH concentrations ( $df=19$ ,  $r^2=0.41$ ,  $p=0.002$ ). In contrast, distance to highway was not significantly related to PAH concentrations, even when standardized by SOM content (Fig. 5), despite evidence that vehicle emissions from the highway were the primary source.



**Fig. 3** Relationship between  $\sum 16$ PAHs and highway traffic across all sites in the Phoenix metro area;  $N=60$ . Each point represents one sample consisting of three homogenized soil cores, showing y-axis error bars for analytical error



**Fig. 4** Relationship between  $\sum 16$ PAHs and SOM across all highway sites in the Phoenix metro area;  $N=60$ . Each point represents one sample consisting of three homogenized soil cores, showing bidirectional error bars for analytical error

## 4 Discussion

### 4.1 Fate of PAHs in Highways of Arid Urban Soils: Relative Importance of Sources, Soil Characteristics, and Climate

Although the profile of PAH compounds in soils varies depending on the type of sources, their magnitude and distribution is frequently explained by source-based hypotheses, as concentrations have been shown to be positively related to the number or proximity of pollutant producers (Johnsen and Karlson 2007; Wang et al. 2008; Wang et al. 2009; De La Torre-Roche et al. 2009). Anthropogenic emissions and deposition of atmospheric compounds generally follow urban–rural gradients, with decreasing PAH concentrations found in soils from industrial, roadside, urban, residential, sub-urban, to rural regions, respectively (Masih and Taneja 2006; Cai et al. 2008; Maliszewska-Kordybach and Klimkowicz-Pawlas 2009; De La Torre-Roche et al. 2009). Similarly, even though emissions of PAHs from manufacturing activity, cooking industry, forest fires, and other incomplete combustion processes may diffuse widely through the atmosphere and deposit near roads, concentrations of these compounds near highways are expected to be vehicle-derived and have been shown to decline exponentially with distance from traffic emission sources (Yang et al. 1991; Zehetner et al. 2009). Because sampling in the current study was near highways, it was not

**Table 3** Results from multiple linear regression analysis of soil PAH concentrations (dependent variable) and uncorrelated soil and roadway characteristics (independent variables)

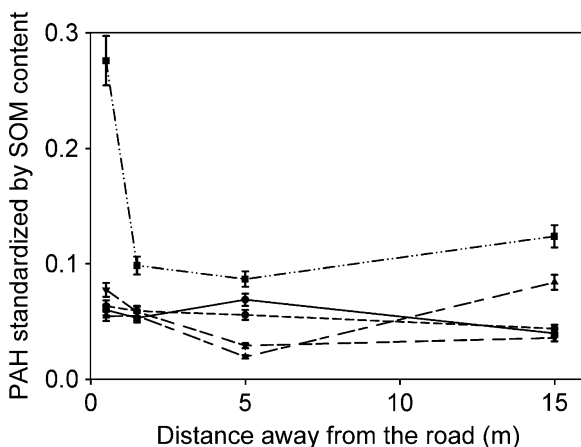
	<i>t</i>	Significance	Standardized $\beta$
<b>Soil organic matter</b>	3.0	<0.01	0.55
<b>Traffic volume</b>	2.7	0.01	0.31
Ammonium	1.6	0.13	
Potential net N mineralization	0.9	0.36	
Water holding capacity	-0.9	0.38	
Potential net nitrification	-0.7	0.46	
Soil moisture	-0.4	0.70	
Nitrate	-0.4	0.73	
Highway age	0.3	0.77	
pH	0.0	0.97	

Significant variables shown in bold, with standardized coefficients ( $\beta$ ).  $N=60$

surprising to find that PAH compounds originated mainly from diesel engines and vehicle fuel burning (Fig. 2; Yang et al. 1991; Rogge et al. 1993). However, our results showed that source-related factors such as traffic volume (Fig. 3), distance away from the road (Fig. 5), or highway age were less related to PAH concentrations than soil characteristics, particularly SOM (Fig. 4). Moreover, the total magnitude of PAHs in Phoenix highway soil was relatively low despite high vehicle traffic loads compared with other cities (Table 2).

Volatile and semi-volatile chemicals in the environment deposit to surfaces or volatilize in order to

reach equilibrium between the soil and atmosphere (Kaupp and McLachlan 1999; Bozlaker et al. 2008). This equilibrium is affected by PAH concentration, temperature, and various soil properties that influence retention. For example, SOM is thought to play a key role in the environmental partitioning and storage of PAHs due to its aromatic structure and chemical affinity for hydrophobic compounds (Bucheli et al. 2004; Cornelissen et al. 2005). Also, SOM protects PAHs from being photolyzed, chemically oxidized, and microbially degraded (Guieysse et al. 2004; Lima et al. 2005; Ravindra et al. 2008). In support of these relationships, numerous studies have found a positive significant relationship between SOM and PAHs in both background (i.e., rural) soils and those from urban regions (Wilcke and Amelung 2000; Dai et al. 2008; Nam et al. 2009). These surveys indicate that SOM is strongly associated with LMW compounds, which may diffuse to remote soils through long-range atmospheric transport (Bucheli et al. 2004; Nam et al. 2008). However, other studies found no relationship between these two variables, highlighting the multi-variate nature of pollutant fate in the environment and the lack of synthetic understanding of controlling factors at the ecosystem scale. In these studies, factors such as distance to source (i.e., stronger effect of deposition patterns than SOM retention), continuous input of fresh contamination (i.e., PAH adsorption has not yet equilibrated with the soil), or other environmental conditions may outweigh soil properties that influence pollutant retention (Yang et al. 1991; Hafner et al. 2005; Zhang et al. 2006; Katsoyiannis 2006; Cai et al. 2007; Nam et al. 2008; Maliszewska-Kordybach et al. 2008; Jiang et al. 2009). Our work shows that



**Fig. 5** PAH concentrations standardized by SOM concentrations in soil at different distances away from the road (0.5, 1.5, 5, and 15 m). Each point represents one sample consisting of three homogenized soil cores per distance, showing y-axis error bars for analytical error; each line represents one of five replicate locations

SOM is significantly related to both LMW and HMW concentrations (separately and total PAHs) in low SOM-content soils of a desert city, even in highway soils located directly adjacent to traffic sources. These results emphasize that soil properties play a large role in the fate of PAH compounds in arid ecosystems that occur on the extreme end of SOM content compared with other, more mesic ecosystems worldwide.

PAH partitioning from the soil to the atmosphere is influenced not only by soil properties, but also by abiotic factors such as temperature and solar radiation. For example, even though SOM content can be high in some tropical soils, total PAH concentrations are generally lower in the warm tropics compared with temperate climates (Wilcke et al. 1999; Aichner et al. 2007; Wilcke 2007; Daly et al. 2007). Moreover, the molecular weight of PAH compounds is negatively related to their temperature-dependent volatility. For instance, in US background soils along a climosequence from Texas to Canada, % NAP (a low-molecular weight PAH) ranges from 15% of total PAH content in warm climates (MAT 20°C) to 35% in cold climates (MAT 0°C; Wilcke and Amelung 2000). Supporting these trends, NAP constituted only 1% of the total profile in Phoenix, likely due to the high air temperatures of this arid urban environment which reach >40°C and remain >24°C during the summer (June, July, and August). Furthermore, the profile of PAH compounds in Phoenix roadway soil was dominated by HMW compounds while the total PAH content was significantly lower than in other published studies of roadway soils globally (Table 2). Other abiotic factors also control pollutant retention in soils; for example, high rates of precipitation in some cities may reduce the measurable content of PAHs in soil due to leaching, runoff, or by triggering microbial transformation of soil chemicals (Table 2; Wilcke et al. 1999; Masih and Taneja 2006; McCrackin et al. 2008). In addition, wet deposition may create a splashing and road-runoff effect, elevating pollutant levels near roadways. Despite the relationships in the literature between PAH concentration and other variables (e.g., SOM, temperature, precipitation, population, and traffic volume), the relative importance of these factors is unclear across studies of roadside soils in cities due to inconsistent data collection across studies. Many studies did not report

SOM data, preventing any meta-data analyses comparing SOM as a driver of PAH concentrations across cities (Table 2). However, our research suggests that PAH content of roadway soils may be influenced by air and/or soil temperature across diverse cities and particularly by SOM content within cities of arid climates.

## 5 Conclusions

Soils near highways in the Phoenix metro area experience high traffic loads and contain PAH compounds primarily from automobiles. However, PAH concentrations in roadside soils are low relative to temperate cities and are related more strongly to SOM than source inputs. Globally, PAH fate is explained by a suite of environmental and source factors that vary widely in different regions of the world. Characteristics of dryland ecosystems, including low SOM, high temperatures, and high solar radiation, likely contribute to pollutant transformation and partitioning out of the arid soil environment. Consequently, ecosystems with high temperatures may sustain air pollution in the gas phase, decreasing the sink into soils and increasing the environmental risk for human exposure via the atmosphere (Kaupp and McLachlan 1999; Bozlaker et al. 2008). Additionally, given that the C content of desert and xeric soils is typically low, anthropogenic inputs may serve as stressors as well as resources to C-starved heterotrophic microorganisms with potentially large consequences for soil microbial communities (Hall et al. 2009; Johnsen et al. 2006; Schimel et al. 2007; Semple et al. 2007). Continued urbanization will likely increase PAH source input and modify land cover (e.g., low-organic matter, xeric soil transformed to high-organic matter, mesic lawn), increasing the capacity for pollutant storage. These changes to soil properties will play a large role in affecting PAH loss and retention in arid ecosystems.

**Acknowledgments** We would like to thank James Hutchings, Tejkaran Dhillon, David Huber, Yun Ouyang, Jolene Trujillo, Elizabeth Cook, Dana Nakase, and Ferran Garcia-Pichel for the training, field/lab assistance, and/or discussion and review of the manuscript. This material is based upon the work funded by the National Science Foundation under Grant No. DEB-0423704, Central Arizona–Phoenix Long-Term Ecological Research. Additionally, this research was supported by the ASU Graduate & Professional Student Association (GPSA) Research Grant.

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