

# Source, profile, and carcinogenic risk assessment for cohorts occupationally exposed to dust-bound PAHs in Lahore and Rawalpindi cities (Punjab province, Pakistan)

Atif Kamal · Riffat Naseem Malik · Tania Martellini · Alessandra Cincinelli

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**Abstract** In this study, the occurrence and concentrations of PAHs in dust samples, which were collected from the traffic police cabins/working spots, located on the main busy roads of Lahore (TP-L) and Rawalpindi (TP-R) cities (Punjab province, Pakistan) and from the public transport vehicles in the same cities (named as AM-L and AM-R, respectively) were determined. The mean and median concentrations of  $\Sigma$ PAHs were observed in the following descending order: AM-L (1340 and 774 ng g<sup>-1</sup> d.w., respectively), TP-L (866 and 854 ng g<sup>-1</sup> d.w.), TP-R (504 and 379 ng g<sup>-1</sup> d.w.), and AM-R (393 and 290 ng g<sup>-1</sup> d.w. Source apportionment of the soil/dust-bound PAHs was also performed. In all the sampling areas, the diagnostic ratios, principal component analysis (PCA) followed by multiple linear regression (MLR) analysis indicated signatures of mixed sources of PAHs (including vehicular exhaust emission, gasoline/diesel, and coal/wood combustion). Estimated results of incremental lifetime cancer risk (ILCR) indicated that the traffic policemen and drivers, especially those in the urban area of Lahore, were at high risk of cancer via routes of dust ingestion and dermal contact. The results of this research could be very useful for the local Government in order to control the exposure and promote

actions to alleviate PAH contamination and to manage health both at work places in the big cities of Pakistan.

**Keywords** Traffic pollution · PAHs · ILCR · Exposure · Pakistan

## Abbreviations

AM-L	Vehicular dust samples collected from the public transport of Lahore city
AM-R	Vehicular dust samples collected from the public transport of Rawalpindi city
ATSDR	Agency for toxic substance and disease registry
IARC	International agency for research on cancer
MW	Molecular weight
PAHs	Polycyclic aromatic hydrocarbons
TP-L	Dust sample collected from the traffic police workplaces in Lahore city
TP-R	Dust samples collected from the traffic police workplace in Rawalpindi city
U.S. EPA	United States Environmental Protection Agency
WHO	World Health Organization

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A. Kamal (✉) · R. N. Malik (✉)  
Environmental Biology and Ecotoxicology Laboratory,  
Department of Environmental Sciences, Faculty of Biological  
Sciences, Quaid-i-Azam University, Islamabad 45320, Pakistan  
e-mail: dr.atifkamal@yahoo.com  
e-mail: r\_n\_malik2000@yahoo.co.uk

T. Martellini · A. Cincinelli  
Department of Chemistry, University of Florence,  
Via della Lastruccia, 3, 50019 Sesto Fiorentino, FI, Italy

## Introduction

Air pollution is an emerging issue in the big cities of Pakistan. Dust and smoke particles in Pakistan are generally twice the world average and five times higher than in the developed countries. The major sources are the accelerated growth in urbanization, due to people migrating to cities, together with the growth of infrastructures, increasing industrial activities without adequate air emission treatment or control, use of ozone-depleting substances, transportation vehicles, and traffic congestion. The increasing number of vehicles in Pakistan

goes side by side with the increasing population, and according to an estimate, ~100 % increase in motor vehicles in Pakistan was observed during period 1990–2005 (Shabbir and Ahmad 2010 and reference therein).

Motor vehicle exhaust releases numerous pollutants into the atmosphere more than any other single human activity, and it represents the most serious and rapidly growing environmental problem in Pakistan, in particular, in big cities such as Lahore and Rawalpindi. In fact, the improved technology alone has not been able to outweigh the amount of pollution emitted by both the increased number of new cars as well as old-technology cars present on the road in developing countries like Pakistan (Kamal et al. 2015). Vehicular transport is one of the most important anthropogenic emission sources of PAHs in the country contributing as much as 60 % of the total PAH emissions (Bilal and Ali 2014; Kamal et al. 2014a). In addition to vehicular exhaust, PAHs are also formed by other anthropogenic activities such as smoking, residential heating, power stations, and industrial activities (Cincinelli et al. 2004; Kamal et al. 2014c). In the air, PAHs are distributed between vapor and particulate phases (Cincinelli et al. 2007; Kamal et al. 2014c) since, various studies have evidenced that harmful PAHs with 5,6- aromatic rings are predominantly found in particulates, because of their low volatility and high molecular weights.

Due to their persistence and hydrophobicity, PAHs are enriched in the soil where they are likely to be retained for a long time (Wang et al. 2013). Thus, soil is usually considered as a main reservoir for PAHs. In the rural and urban environments, the dry and wet depositions facilitate the PAHs accumulation in soil surface and dust (Wild and Jones 1995; Murakami et al. 2005; Yang et al. 2014). It is also known that human exposure to PAHs is higher through soil than air and water in the outdoor environments (Menzie et al. 1992). Settled dust in the indoor and outdoor environments is a mean of non-dietary intake, of PAHs, (Kamal et al. 2014c and reference therein), because PAHs esp. those having high molecular weight are largely adsorbed onto dust particles (Naspinski et al. 2008). Several studies have reported the concentration of PAHs in surface dust (Liu et al. 2007; Dong and Lee 2009; Kamal et al. 2014c). PAHs in soil/dust phase persist for a longer period of time and thus serve as a tool for assessing the health risk to the exposed population (Wang et al. 2013; Kamal et al. 2014c).

Exposure to PAH is associated with several adverse human health effects such as hematological changes/anemia (Kamal et al. 2014b, c), genotoxicity, hepatotoxicity, lung cancer, neurological dysfunction, and bladder cancer (Kamal et al. 2014a, b, c and references therein). In response to the worsening traffic scenario present in big cities, the occupational environment may be the most important component of the exposure of traffic-related workers to PAHs. In the last decade, few studies have evidenced the high exposure to PAHs among traffic policemen and, road builders, in Budapest (Hungary) (Szaniszlo and Ungvary 2001), Bangkok (Thailand)

(Ruchirawa et al. 2002), and Beijing (China) (Liu et al. 2007), focusing, however, their attention only on the determination of PAHs in the air particulate and gas phases. A recent study conducted in Pakistan has shown that human cohorts, highly exposed to PAHs from traffic pollution, suffer from significant oxidative stress due to continual exposure during outdoor work (Kamal et al. 2015). Further studies evidenced the health risks for brickkiln workers in contact with dust-bound PAHs (Kamal et al. 2014a) and highlighted that in-vehicle ambient is one of the most contaminated microenvironments (Abdallah et al. 2008). Aims of this study were to determine the occurrence and levels of PAHs in soil/dust samples collected in public transport vehicles and traffic police cabins/working spots in Pakistan, to assess source contributions to the soil/dust PAH burden and to evaluate the incremental lifetime cancer risk (ILCR) via inhalation, dermal contact, and ingestion exposure. The results of this research may be very useful for the local Government in order to control the exposure and promote actions to alleviate PAH contamination and occupational health management.

## Materials and methods

### Chemicals and reagents

All solvents were pesticide grade and purchased by Supelco (Bellefonte, PA, USA) and tested for contaminants before use. Standard PAH mixture EPA was purchased from Supelco (Bellefonte, PA, USA). Benzo(e)pyrene and coronene were purchased from Alltech (Deerfield, USA). Silica (100–200 mesh) and sodium sulfate ( $\text{Na}_2\text{SO}_4$ ) were purchased from Merck (Darmstadt, Germany). Sodium sulfate was heated for 12 h at 450 °C to remove any organic matter and kept at 120 °C until use.

### Sample collection and processing

It is worth describing that collection of pure dust was often not possible in most of the cases, since, the dust samples were always accompanied by some fractions of soil particles, therefore, the dust samples in this section are described as “soil/dust samples”, instead of “pure dust particles”. The samples were collected from the public transport vehicles and traffic police cabins/working spots located in high-traffic density roads in the cities of Lahore and Rawalpindi, located in the Punjab province of Pakistan. Lahore, is the second-largest city of Pakistan and is inflicting in swelling air pollution mainly attributed to motor vehicle emissions (Schneidmesser et al. 2010). Rawalpindi is the fourth most popular urban area, and it is a rapidly growing city in the Potohar region of the northern Punjab.

Soil/dust samples were collected using dustpans and plastic brushes (with sterile bristles). We always used a new brush and dustpan for each sampling site to avoid cross-contamination. A total of 21 soil/dust samples (~20 g) were collected from traffic police working spots (areas occupied by the traffic police to control traffic, TP) in Lahore and Rawalpindi. For each sampling site, 6–8 subsamples of soil/dust were collected from the area of 1 m<sup>2</sup> and bulked together to form one composite sample. The soil/dust samples ( $n=20$ ) from the interior of the public transport vehicles (i.e., Hiace vegans and busses) were also collected from the same cities. Only Hiace vegans (vehicles having a capacity of carrying 16–18 passengers) were available to commuters on the daily basis in Rawalpindi city whereas mini and large busses and vegans operated routinely in Lahore city. A large number of busses and vegans in Lahore city are very old (i.e., manufactured before 2000). Drivers of selected busses and vegans were asked not to wash/vacuum their vehicles for at least a week (preferably 2 weeks) before sample collection. Samples were collected from each vehicle for two consecutive days at the end of the work-shift from three different locations inside the vehicle: dashboard and driver cabin (including steering, dashboard panel, and armrest levels), passenger cabin, and rear parts of the vehicle. After collection, samples were homogenized into one composite sample. In order to protect all samples from sunlight exposure, aluminum foil (pre-rinsed with solvents) was used to wrap each sample separately. After that, the samples were sealed in zip-locked polyethylene bags and stored until analysis.

Samples were classified as AM-L (vehicular dust samples from Lahore city), AM-R (vehicular dust samples from Rawalpindi city), TP-L (traffic policemen working areas in Lahore city), and TP-R (traffic policemen working areas in Rawalpindi city) (see Fig. 1).

#### Sample preparation, extraction, cleanup, and analyses

The details of sample preparation and instrumental analyses have been described in detail previously (Martellini et al. 2012; Kamal et al. 2014a). Briefly, 3 g of soil/dust sample was homogenized and was spiked with a mixture of deuterated standards (*p*-terphenyl-d<sup>4</sup>, fluoranthene-d<sup>10</sup>, and benzo(a)pyrene-d<sup>12</sup>). The extraction procedure was carried out in an ultrasonic bath (using crushed ice for pre-cooling) for 15 min, using a solution of hexane and dichloromethane (1:1 v/v). After this step, the extracts were vortex stirred (1 min) and centrifuged for 5 min at 2500 rpm. The supernatants were reduced approximately to 1 ml volume under a gentle stream of ultrapure nitrogen. The concentrated extract was cleaned up on a silica gel column. The column was first eluted with 25 ml of hexane and then with 50 ml of a mixture of hexane and dichloromethane (4:1 v/v). The resulting extracts were concentrated to 1 ml volume under the gentle

stream of ultrapure nitrogen, and transferred into a vial for storage. Before analysis, the extracts were spiked with an internal standard mixture containing naphthalene-d<sup>8</sup>, phenanthrene-d<sup>10</sup>, chrysene-d<sup>12</sup>, and indeno(1,2,3-cd)pyrene-d<sup>12</sup>. The samples were injected into a Hewlett–Packard 6890 gas chromatography–mass spectrometer (GC–MS), equipped with a 5973 mass selective detector (MSD), and an HP-5MS capillary column (J&W Scientific, Folsom, CA, USA; 30 m, 0.25 mm I.D., 0.25 mm film thickness). Compound identification was based on the MSD database (NIST, 98) and GC retaining time of authentic PAH standards. The MSD was operated in selected ion monitoring mode (SIM).

#### Quality control and quality assurance (QC/QA)

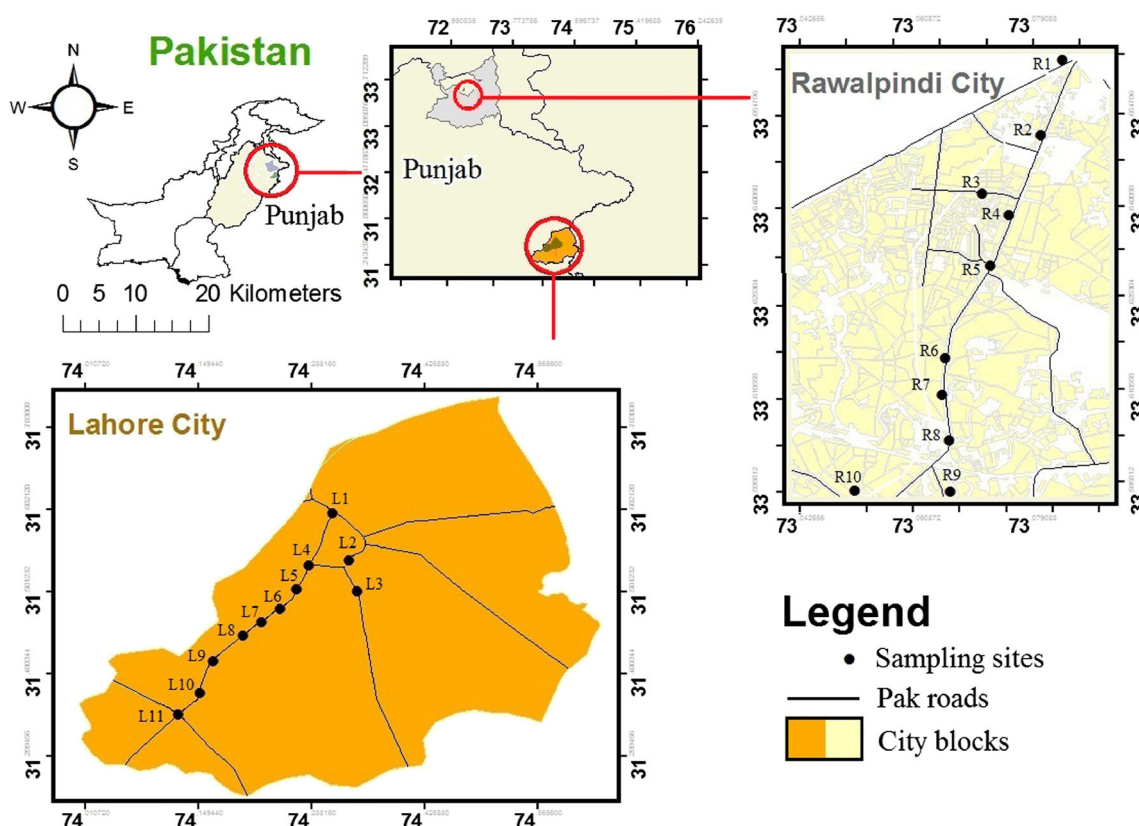
Average PAH recoveries and relative standard deviation (RSDs) were measured to evaluate the method performance by multiple analyses of clean sand samples spiked with PAH standard mixture containing 18 PAHs (acenaphthene (Ace), acenaphthylene (Acy), anthracene (Ant), benzo(a)anthracene (BaA), benzo(a)pyrene (BaP), benzo(b)fluoranthene (BbF), benzo(e)pyrene (BeP), benzo(ghi)perylene (BghiP), benzo(k)fluoranthene (BkF), chrysene (Chry), coronene (Cor), dibenzo(a,h)anthracene (DBA), fluoranthene (Fla), fluorene (Fl), indeno(1,2,3-cd)pyrene (IP), naphthalene (Naph), phenanthrene (Phe), pyrene (Pyr). BbF and BkF were reported together as BbkF. In order to determine any potential laboratory contamination, the procedural blanks were run periodically. The recovery percentage of 18 PAHs ranged between 84.1 and 122.4 %, while that of surrogate PAHs was 93±13 % for benzo(a)pyrene-d<sup>12</sup>, 97±12 % for fluoranthene-d<sup>10</sup>, 94±11 % for *p*-terphenyl-d<sup>4</sup>. We also used SRM1649a (urban dust) as a control. The diluted standards used were between the range of 0.16 and 10 ng μl<sup>-1</sup>, and a six-point calibration curve was drawn for quantification of PAHs in the soil/dust samples. Detection limits of PAHs were in the range of 20–60 pg g<sup>-1</sup> d.w. (dry weight basis).

#### ILCR assessment/model and parameters

The exposure risk of workers (drivers and traffic policemen) to dust-bound PAHs was estimated using the probabilistic incremental lifetime cancer risk (ILCR) based on the U.S. EPA standard risk assessment models (U.S. EPA 1991; Table 1).

The exposure risk was calculated only for adults, taking into account that age of the transport workers ranged between 18 and 70 years. The ILCR in terms of direct ingestion, dermal contact, and inhalation after exposure to dust-bound PAHs were calculated as follows (please see ESM1 for the details of models used for ILCR calculation):

where, ABS is the absorption factor, AF the adherence factors, AT the average lifetime, BW the recommended body weight of the exposed person, CF the conversion factor, CSF



**Fig. 1** Geographical locations of the sampling sites in the Punjab province (Pakistan)

the carcinogenic slope factor (based on the cancer-causing ability of benzo(a)pyrene), Conc. Dust; the concentration of the chemical of interest in dust, ED the exposure duration, EF the exposure frequency, *IngR* the ingestion rate, *InhR* the inhalation rate, PEF the particle emission factor for BaP, SA the workers exposed skin surface area, and  $\Sigma$ TEQ the sum of BaP equivalent concentration of PAHs. The details of values and units of parameters used in these formulae are reported in Table 1 and were based on the risk assessment guidance of U.S. EPA and related publications.

**Coefficient of divergence**

The coefficient of divergence (CD) was used to identify the similarities between the profiles of PAHs at two different sites (e.g., city-wise comparison). The generic formulae used for CD calculation were as follows: (also see ESM1)

$$CD_{jk} = \sqrt{\frac{1}{P} \sum P_i = 1(X_{ij} - X_{ik} X_{ij} + X_{ik})^2}$$

The *j* and *k* represent the profile of PAHs in two different sampling sites, *p* shows the number of target compounds, and *x* is the average concentration of the component

(Wongphatarakul et al. 1998). The values of CD <0.269 reflect a probable divergence between particles of two sites (cities in this case); a CD value >0.16 suggests a common source in two sites under discussion (Wongphatarakul et al. 1998).

**Statistical analyses**

Individual and total PAHs concentrations detected in dust samples and the molecular diagnostic ratios calculated for source identification have been presented in mean ± standard deviation (Mean ± S.dev), median (Med), minimum (Min), and maximum (Max) values. All the PAHs results were presented on the basis of dry weight. Normality of data was tested with the Kolmogorov–Smirnov (K-S) normality test. Principal component analysis (PCA) followed by the multiple linear regression (MLR) was also used for source identification using varimax rotation and extracting principal components having eigenvalues >1. Differences between sites were assessed using Mann–Whitney’s *U* test. The benzo(a)pyrene equivalent concentrations (BaP.eq.) were determined using the toxic equivalency factor (TEF) provided by Nisbet and LaGoy (1992), U.S. EPA (U.S. EPA 1989, 2005, 2009), and Malcom and Dobson (1994). All statistical analyses were performed using SPSS software (version 20) for windows.

**Table 1** Parameters used to estimate ILCR for workers exposed to dust-bound PAHs

Model parameters	Abbreviations	Previous default	Currently recommended	Unit	References
Adult/child soil ingestion rate	IngR	Outdoor workers=100	–	mg day <sup>-1</sup>	U.S. EPA 1991 pg. 15
Average lifetime days	AT(days)=70 years×365 day <sup>-1</sup>	25,550	–	Days	U.S. EPA 2009
Body weight of the exposed person	BW	Adults=70	Adult=80	kg	U.S. EPA 1991; <sup>a</sup> U.S. EPA 2011a, Table 8–3; weighted mean values for adults 21–78
Chemical concentration in dust	Conc. Dust	Study specific	–	ng g <sup>-1</sup> d.w.	This study
Conversion factor	CF	1×10 <sup>-6</sup>	–	–	U.S. EPA 1989
Dermal absorption factor	ABS	0.13	–	–	Man et al. 2013
Worker soil adherence factors	AF soil	Adults=0.2	0.12	mg/cm <sup>2</sup>	U.S. EPA 2002; Exhibit, 1–2; U.S. EPA 2011a <sup>a</sup> , Table 7–20 and Section 7.2.2
Worker soil exposure time	ET	8	8	h day <sup>-1</sup>	U.S. EPA 1991, 2013a, b (pg. 15)
Outdoor worker exposure duration	ED	25	–	Years	U.S. EPA 1991, 2013a, b (pg. 15)
Outdoor exposure frequency	EF	313 (working days)	–	Days year <sup>-1</sup>	Based on survey in the current study
Particle emission factor for BaP	PEF	1.36×10 <sup>9</sup>	–	m <sup>3</sup> kg <sup>-1</sup>	U.S. EPA 2013a, b
Workers soil surface area exposure	SA	3300	3470	cm <sup>2</sup>	U.S. EPA 2002 (Exhibit 1–2) U.S. EPA 2011a <sup>a</sup> , Table 7–2;
Carcinogenic slope factor	CSF	CSF <sub>ingestion</sub> =7.3 CSF <sub>dermal</sub> =25 CSF <sub>inhalation</sub> =3.85	–	(mg kg <sup>-1</sup> day <sup>-1</sup> ) <sup>-1</sup>	Peng et al. 2011
Inhalation rate	InhR	Adult=20	–	m <sup>3</sup> day <sup>-1</sup>	SFT 1999
Cancer risk estimate	CR	≤10 <sup>-6</sup> =low 10 <sup>-4</sup> ≤to <10 <sup>-3</sup> =moderate 10 <sup>-3</sup> ≤to <10 <sup>-1</sup> =high ≥10 <sup>-1</sup> =very high	–	–	NYSDOH et al. 2007

<sup>a</sup> Reference for new recommendations; other values are either not changes or remained the same in new recommendations by EPA

## Results and discussion

### Profile of PAHs in soil/dust samples

The profile of PAHs (individual and  $\Sigma$ PAHs concentrations) in surface dust samples is summarized on the basis of sampling sites in Table 2. The overall profile based on molecular weight of PAHs was almost similar in dust samples from TP and AM, with relatively high contents of low and high molecular weight PAHs in dust samples of TP worksites. The mean and median concentrations of  $\Sigma$ PAHs were observed in the following descending order: AM-L (1340 and 774 ng g<sup>-1</sup> d.w. respectively), TP-L (866 and 854 ng g<sup>-1</sup> d.w.), TP-R (504 and 379 ng g<sup>-1</sup> d.w.), and AM-R (393 and 290 ng g<sup>-1</sup> d.w.). Highest mean concentrations were observed in AM-L and

TP-L samples ranging from 385 to 3539 ng g<sup>-1</sup> d.w. and from 357 to 4530 ng g<sup>-1</sup> d.w., respectively. The  $\Sigma$ PAHs of TP-L sites in this study were higher than the average  $\Sigma$ PAHs reported in the street dust samples of China by Jiang et al. (2014) (i.e., 3900 ng g<sup>-1</sup>), ~Chung Khurd (882±167 ng g<sup>-1</sup> d.w.) Pakistan, but lower than those measured in Sohdra town (2578 ng g<sup>-1</sup> d.w.) and Gujranwala city (957 ng g<sup>-1</sup> d.w.) (Kamal et al. 2014a). The values were also higher than the reported mean  $\Sigma$ 16-PAH concentration in Korean rural areas (220 ng g<sup>-1</sup> d.w.), urban areas (390 ng g<sup>-1</sup> d.w.), and industrial areas (1900 ng g<sup>-1</sup> d.w.) (Kwon and Choi 2014). The average concentration of  $\Sigma$ PAHs in AM-L samples was also higher than that found in urban dust samples of Bangkok (1.1±0.801 μg g<sup>-1</sup>) (Boonyatumanond et al. 2007), Dalian, China (1.89–17.1 μg g<sup>-1</sup>) (Wan et al. 2006), and Guangzhou (China)

**Table 2** Summary of PAHs detected in soil/dust samples collected from the selected occupational sites in Rawalpindi and Lahore cities of Punjab province (Pakistan)

PAHs ng g <sup>-1</sup> d.w. PAHs (rings #) (abbreviations) /TEFs	TP-R (10)			TP-L (11)			AM-R (10)			AM-L (10)		
	Mean±S.dev	Med	Min-Max	Mean±S.dev	Med	Min-Max	Mean±S.dev	Med	Min-Max	Mean±S.dev	Med	Min-Max
Naphthalene (3) (Naph) 0.001	11.9±3.7, 8.8	8.8	3.55–28.6	22.5±9.5, 10.97	10.97	1.58–88	0.56	7.33±1.3, 5	5	3.6–14.8	11.7	7.8–69.3
Acenaphthene (3) (Ace) 0.001	14.2±6.8, 16.4	16.4	3.75–27.6	28±9, 22.3	22.3	2.99–115	0.17	13.9±8, 9.8	9.8	7.66–33	22.3	5.27–51.9
Acenaphthylene (3) (Acy) 0.001	10.7±2.3, 11.4	11.4	2.85–21.6	11.8±4.7, 11	11	2.2–26.8	0.69	10.6±3.6, 9.8	9.8	7–15	16.3	6.6–96.7
Anthracene (3) (Ant) 0.01	17±4.8, 15.5	15.5	4.74–30.2	52.8±49, 26.43	26.43	3.72–130	0.24	12.7±2.7, 12.98	12.98	11.8–13.2	38	18.9–135
Fluorene (3) (Fl) 0.001	17.1±8.7, 18.1	18.1	4.45–32.8	33.1±37, 20	20	3.46–136	0.43	16.5±7, 14	14	8.67–30.5	28.7	23–75.5
Phenanthrene (3) (Phe) 0.001	48.2±8, 40.9	40.9	12.6–93.2	141±56, 65	65	8.52–903	0.27	32±8, 34.7	34.7	23–41.3	106.8	34.9–407
Benzo(a)anthracene (4) (BaA) 0.1	22.7±12, 14.6	14.6	5.1–39.5	44.5±8, 25	25	2.22–285	0.6	20.5±4.6, 10.9	10.9	10–46	34.8	17.7–205
Chrysene (4) (Chry) 0.01	59.2±3.8, 43.6	43.6	20–112.8	61.95±5.6, 47.7	47.7	3.98–309	0.33	23.4±4.7, 21.6	21.6	17.9–31	79.5	33.6–424
Fluoranthene (4) (Fla) 0.001	60±35, 54.5	54.5	14.8–109	123.7±28, 66.6	66.6	7.05–800	0.89	37±8, 28	28	16–62.7	101	27.8–613
Pyrene (4) (Pyr) 0.001	49.2±10, 43.42	43.42	12.6–85.3	98.9±15.8, 63	63	7.04–590	0.94	25.8±24, 15.9	15.9	17.9–273	72	33.4–423
Benzo(a)pyrene (5) (BaP) 1	29.7±16, 20	20	8.61–55.4	46.7±9, 25	25	6.87–275	0.47	25.8±24, 15.9	15.9	8.6–71	31.2	18.3–171
Benzo(b+k)fluoranthene (5) (BbKf) 0.1	47.5±11, 32	32	10.7–88	46.5±3.9, 25.7	25.7	2.16–255	0.24	13.4±3, 13	13	9.7–18.9	46	17.8–323
Dibenzo(a,h)anthracene (DBA) 1 (5)	10.8±3.8, 9.72	9.72	2.64–21.6	13±8.25, 15	15	1.47–27.6	0.6	11.6±5.4, 13	13	5.14–18.6	41	12.9–117
Benzo(e)pyrene (5) (BeP) 0.01	28.9±6.9, 18.5	18.5	7.1–51	34.5±6.7, 28	28	2.4–168	0.47	20.1±6.7, 12.8	12.8	10.9–51.8	28	20.8–135
Indeno(1,2,3-c,d)pyrene (6) (IP) 0.1	27.2±6.6, 20.4	20.4	6.92–49.6	26.2±7, 19	19	1.6–110	0.36	25.8±3, 15.9	15.9	7.1–68	29	21.7–166
Benzo (g,h,i)perylene (6) (BghiP) 0.01	49.7±8, 37.6	37.6	15.8–85.6	45.5±9.4, 44	44	4.44–121	0.51	49.9±15, 25.8	25.8	17.7–133	57.5	41.5–215
Coronene (7) (Cor) 0.001	33±5, 33	33	9.67–57.2	24.7±2, 19.8	19.8	4.27–73.3	0.32	40.6±8, 21	21	14.7–104	53	40.5–123
Ant/(Phe+Ant)	0.27±0.2, 0.27	0.27	0.23–0.34	0.28±0.12, 0.28	0.28	0.22–0.3	0.33	0.29±0.1, 0.27	0.27	0.22–0.37	0.26	0.23–0.38
BaP/(BaP+Chry)	0.27±0.13, 0.49	0.49	0.45–0.53	0.5±0.2, 0.53	0.53	0.38–0.58	0.01	0.46±0.2, 0.45	0.45	0.32–0.57	0.47	0.41–0.59
BaP/BghiP	0.61±0.34, 0.62	0.62	0.47–0.92	0.72±0.17, 0.48	0.48	0.24–2.28	0.56	0.61±0.12, 0.53	0.53	0.21–1.29	0.54	0.36–1.83
BeP/(BeP+BaP)	0.49±0.25, 0.49	0.49	0.45–0.53	0.49±0.12, 0.53	0.53	0.38–0.58	0.65	0.46±0.23, 0.45	0.45	0.32–0.57	0.47	0.41–0.59
Fl/(Fl+Pyr)	0.26±0.16, 0.26	0.26	0.14–0.38	0.31±0.13, 0.31	0.31	0.17–0.5	0.27	0.32±0.24, 0.33	0.33	0.10–0.49	0.28	0.15–0.50
Fla/(Fla+Pyr)	0.26±0.16, 0.30	0.30	0.20–0.31	0.35±0.16, 0.34	0.34	0.27–0.48	0.21	0.38±0.23, 0.35	0.35	0.28–0.60	0.35	0.29–0.59
IP/(IP+BghiP)	0.35±0.32, 0.35	0.35	0.30–0.38	0.32±0.14, 0.28	0.28	0.25–0.48	0.06	0.35±0.12, 0.34	0.34	0.25–0.47	0.34	0.31–0.44
Phe/(Phe+Ant)	0.73±0.34, 0.73	0.73	0.66–0.77	0.72±0.12, 0.72	0.72	0.7–0.78	0.33	0.71±0.1, 0.73	0.73	0.63–0.78	0.74	0.63–0.77
Σ PAHs	504±182, 379	379	136–855	866±159, 854	854	357–4550	0.94	393±54, 290	290	2340–872	774	385–3539
Σ4,5,6,-rings PAHs/ΣPAHs	0.76±0.33, 0.76	0.76	0.68–0.83	0.63±0.32, 0.62	0.62	0.56–0.81	<0.01	0.7±0.02, 0.66	0.66	0.6–0.86	0.68	0.6–0.87
Σ 7-Carcinogenic PAHs	226±130, 168	168	59.7–389	308±58, 213	213	19.4–1638	0.56	207±50, 113.9	113.9	83.6–604	298	154–1502
Σ COMB	374±93, 283	283	102–635.6	528±77, 356	356	32.8–2914	0.05	288±38, 177	177	145–738	549	249–2677
ΣCOMB/ΣPAHs	0.74±0.32, 0.74	0.74	0.65–0.81	0.61±0.32, 0.59	0.59	0.54–0.79	<0.01	0.68±0.33, 0.67	0.67	0.59–0.85	0.64	0.55–0.76
ΣLMPAHs/ΣLMPAHs	0.31±0.12, 0.31	0.31	0.21–0.47	0.7±0.26, 0.77	0.77	0.28–0.96	<0.01	0.56±0.28, 0.57	0.57	0.21–0.97	0.63	0.35–0.94

Coefficient of divergence (CD) values=0.227 (for TP-R and AM-R), and 0.257 (for TP-L and AM-L); TEF for individual PAHs relative to BaP as reported by Nisbet and LaGoy (1992), except TEF values for Cor and BeP which were adopted from Malcolm and Dobson (1994). P=probability value, significant when <0.05; ΣCOMB=PAHs of combustion origin, i.e., BaA, BaP, BbKf, BeP, BghiP, Chry, Fla, IP, Pyr (Prahl and Carpenter 1983); Σ7-Carcinogenic PAHs=BaA, BaP, BbKf, BghiP, IP, Pyr  
 ΣPAHs sum of non-alkylated PAHs, LMPAHs low molecular weight PAHs, HMPAHs high molecular weight PAHs; a= comparative analysis of PAHs between TP-L and TP-R dust samples; b= comparative analysis of PAHs between AM-L and AM-R dust samples

(0.84–12.3  $\mu\text{g g}^{-1}$ ) (Wang et al. 2011a, b) and comparable to those found in the northeast England (ranging between 0.60 and 46  $\mu\text{g g}^{-1}$ ) (Lorenzi et al. 2011).

The average  $\Sigma$ 7-carcinogenic PAHs values were 530 and 308  $\text{ng g}^{-1}$  d.w. in AM-L and TP-L samples, respectively, whereas 207 and 226  $\text{ng g}^{-1}$  d.w. were determined in AM-R and TP-R samples, respectively (Table 2), accounting for 36–53 % of total PAHs. Among the carcinogenic PAHs, like Chry, Fla, BghiP, and Pyr were the most abundant in most of the dust samples. The average concentration of BaP was higher in soil/dust samples of Lahore as compared to that of Rawalpindi city. The AM-L samples showed higher mean concentration of BaP (71.8  $\text{ng g}^{-1}$  d.w.) than the detected BaP concentration in the samples of TP-L worksites (46.7  $\text{ng g}^{-1}$  d.w.).

The average concentration of  $\Sigma$ combustion origin PAHs ( $\Sigma$ COMB) showed the highest value in AM-L samples (917  $\text{ng g}^{-1}$  d.w.). Comparatively, the average concentration of almost all the individual PAHs was significantly higher in dust samples of AM-L as compared to that of AM-R ( $P < 0.05$ ); however, no significant differences were observed between PAH congeners in TP-L and TP-R soil/dust samples ( $P > 0.05$ ), where the average concentration of individual PAHs was higher in TP-L as compared to that of TP-R. Moreover, the PAH concentrations remained generally higher in soil/dust samples collected from the interior of the vehicles than in those collected from the traffic police working spots.

As expected, overall the individual PAH concentrations were higher in dust samples of Lahore than Rawalpindi, indicating a more remarkable contribute of heavy traffic and human activities in this city. In fact, even if Rawalpindi is adjacent to Islamabad city (31° 28' 50" North, 74° 24' 50") and influenced by different anthropogenic activities, in particular transport sector, it is considered less polluted than Lahore. Lahore is a very old and overpopulated city as compared to Rawalpindi, with a huge number of public transport vehicles and consequently higher level of traffic pollution. Moreover, the proximity of Lahore to the Grand Trunk (GT) road of Pakistan and the presence of a large number of industrial activities and brick manufacturing units in its suburbs together with the use of wood and coal for residential cooking purposes in the small villages close to the city, contribute to the air pollution of the city and the high PAH levels in soil/dust samples. In a recent study, it has been documented that Lahore is afflicted with high levels of particle air pollution well above the guidelines of the World Health Organization (WHO) (Stone et al. 2010); daily ambient concentrations of  $\text{PM}_{2.5}$  (monthly average  $200 \pm 80 \mu\text{g m}^{-3}$ ) and  $\text{PM}_{10}$  (average  $340 \pm 100 \mu\text{g m}^{-3}$ ) exceeded the WHO guideline values (WHO 2008) i.e 25 and  $50 \mu\text{g m}^{-3}$ , respectively, showing higher airborne PAH levels than those found in other polluted Asian cities like Hong Kong and Mumbai (Stone et al. 2010). The same authors (Stone et al. 2010) found high organic carbon content in the atmosphere of Lahore due to the influence of anthropogenic activities like the

combustion of biomass fuels, motor vehicles, and motor oil gasoline combustion in non-catalytic gasoline engines and influence of combustion emission from the brick-making activities.

#### PAH source apportionment

The relative abundances or molecular diagnostic ratios were used to characterize and identify PAH emission sources such as combustion and traffic exhaust emissions. Source identification may provide the origin and contribution of more than one non-point source at particular exposure sites and may be helpful in evaluating the associated risk to both environment and human.

Diagnostic ratios of PAHs can be used for qualitative investigation of the source contribution. The most frequently used diagnostic ratios and the qualitative information on source apportionment of PAHs in some other publications as well as in this study are reported in Table 3, see also electronically submitted material (ESM) 1. In general, PAHs in urban surface dust had a common characteristic with high molecular weight PAH (HMW PAH) (4, 5-rings) contribution (62 % to 94 %), indicating a pyrogenic origin (Zakaria et al. 2002). The overall PAH profile in this study shows abundance of 4-rings followed by 5-rings PAHs, which represent a signature of biomass fuel combustion and purified oil (Robinson et al. 2006; Larsen and Baker 2003). The 2- and 3-ring PAHs, which are probable markers of petroleum-derived residues (Sporstol et al. 1983), were present in lower concentration than 4, 5-rings PAHs showing dominance of overall combustion from biomass fuel exhaust. BbkF could represent the possible contribution of diesel exhaust (Larsen and Baker 2003; Robinson et al. 2006), whereas the elevated BghiP and BaP concentrations in dust samples from Lahore could represent the contribution of vehicular exhaust emission (Harrison et al. 1996; Boonyatumanond et al. 2007). Similarly, IP represented the diesel (Robinson et al. 2006) and gasoline emissions (Boonyatumanond et al. 2007). Generally, the Cor comes from automobile exhaust emissions, therefore, the high concentrations, (ranging between 4.27 and 123  $\mu\text{g g}^{-1}$  d.w) indicated the dominant contribution of this source. Both the general profile of PAHs and diagnostic ratios indicated traffic exhaust emission as a major contributor, with signature of biomass/coal wood combustion. The principal component analysis (PCA) and multiple linear regression (MLR, see Supplimentary file S1) was also used to select statistically independent source tracers and to simplify the interpretation of complex data set. The PC-I and PC-II of TP dust samples marked the contribution of mixed (traffic and biomass combustion activity) and petrogenic (traffic emission) sources, respectively, in traffic hotspots (Fig. 2a). Thus, overall results of PCA of AM samples (Fig. 2b) represented a PAH profile reflecting major contribution of gasoline/petroleum combustion, and the outdoor dust source which contributes to the dust deposition inside vehicles (see ESM 1 for details).

**Table 3** Multiple diagnostic ratios used for the source identification of PAHs in soil/dust samples collected from the traffic police working areas (TP) and public transport vehicles (AM)

PAH diagnostic ratios	Prescribed ranges	TP <sup>a</sup> samples	AM <sup>a</sup> samples	Probable source(s)	References
$\Sigma 4,5,6\text{-rings PAHs}/\Sigma\text{PAHs}$	<0.4=Petrogenic >0.5=Petroleum burn+coal/ softwood combustion	0.7	0.7	Petrogenic/combustion	Biache et al. 2014
$\Sigma\text{COM}/\Sigma\text{PAHs}$	0.3=Petrogenic 0.7=Pyrogenic ~1=Combustion	0.68	0.66	Petrol emissions	Pandey et al. 1999; Ravindra et al. 2008a
$\Sigma\text{LMPAHs}/\Sigma\text{HMPAHs}$	<1=Pyrogenic >1=Petrogenic ~1=Combustion	0.51	0.58	Petrogenic	Zhang et al. 2008
Ant/(Phe+Ant)	<0.1=Petrogenic >0.1=Pyrogenic	0.28	0.29	Petrogenic	Pies et al. 2008
BaP/(BaP+Chry)	<0.2=Petrogenic >0.35=Vehicular, combustion	0.31	0.39	Traces of coal in TP dust samples; vehicular combustion in AM dust samples	Akyuz and Cabuk 2008; Yunker et al. 2002
BaP/BghiP	<0.6=Non-traffic 0.5–0.6=Traffic >1.25=Traffic	0.67	0.7	Traffic	Pandey et al. (1999), Park et al. (2002)
BeP/(BeP+BaP)	~0.5=Fresh particles <0.5=Photolysis	0.5	0.48	Fresh in the case of TP dust samples; photolysis in the case of AM dust samples	Oliveira et al. 2011
Fl/(Fl+Pyr)	<0.5=Gasoline emissions >0.5=Diesel	0.29	0.31	Gasoline emission	Ravindra et al. 2008b; Mandalakis et al. (2002); Fang et al. (2004a, b), Ravindra et al. (2006a, b)
Fla/(Pyr+Fla)	<0.4=Petrogenic 0.4–0.5=Fossil fuel >0.5=Wood/coal/grass	0.53	0.52	Fossil fuel	De La Torre-Roche et al. 2009; Yunker et al. 2002
IP/(IP+BghiP)	<0.2=Petrogenic 0.2–0.5=Petroleum combustion	0.33	0.36	Diesel	Yunker et al. 2002
Phe/(Phe+Ant)	>0.70=Lubricant oil/fossil fuels	0.72	0.71	Fossil fuel	Alves et al. 2001

The symbol tilde (~) indicates roughly similar

<sup>a</sup> Average values

*Coefficient of divergence (CD) analysis*

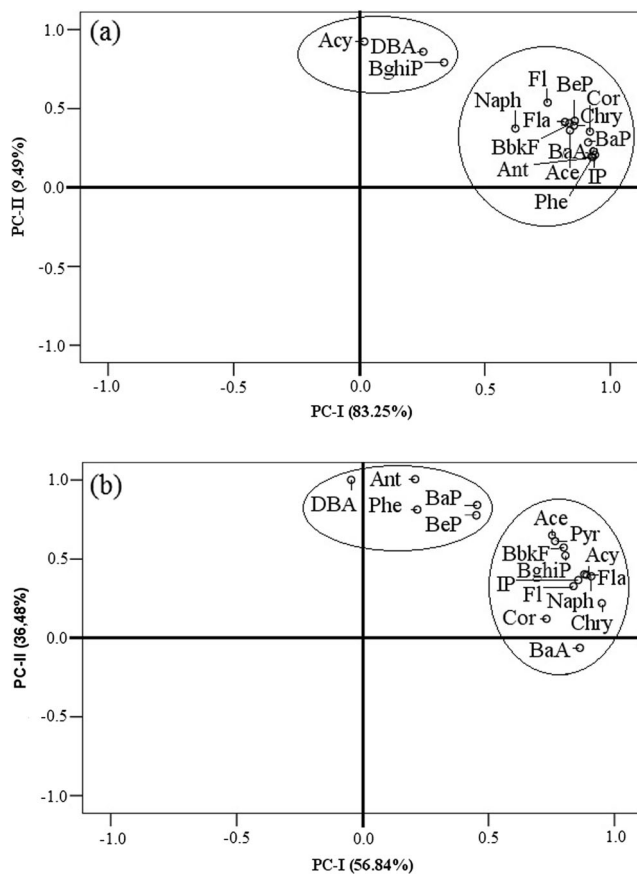
The CD was used for the analysis of similarities or dissimilarities between the dust samples taken from two compartments (indoor AM and outdoor TP worksites). In general, it is supposed that the CD value approaching zero indicates the similarity of source, while according to Wongphatarakul et al. (1998), a CD value of 0.269 may be a reflection of the divergence between particles from two different sites; in this study, we calculated CD values to evaluate the divergence of dust particles between TP and AM sites of the same cities. The CD values of both cities showed that PAHs in different compartments were possibly originated from the same sources (Table 2). As evidenced by the PCA analysis for TP sites these sites were in particular affected by the influence of stationary combustion source around the cities, and vehicular exhaust as mobile source. However, in the case of AM samples, the dust deposition was compartmentalized, where PAHs were

originating from the petroleum/diesel combustion, with a major contribution of outdoor dust.

*Evaluation of carcinogenic risk*

To better our knowledge, this is the first study on the assessment of incremental lifetime cancer risk from dust-bound PAHs among TP and AM drivers in Lahore and Rawalpindi cities of Pakistan (Table 4). We assessed risk of exposure for adults only, taking into account the worst-case scenario because of the severity of occupational exposure and because children are also not representative of these two working classes. The novel aspect of this study is related to the evaluation of incremental lifetime cancer risk assessment to specific working classes compared with the ILCR values reported in literature for the general public in a residential setting. Drivers spend their time in the “in-vehicle” micro-environment; therefore, from an enclosed environment, they can be considered to be under high





**Fig. 2** Principal component analyses of PAH congeners detected in the soil/dust samples collected from (a) traffic police worksites and (b) public transport vehicles

risk. It is also important to take into account that some of the parameters that we used in this evaluation are generalized, while some of them are currently revised by U.S. EPA (Table 1). However, there is always a need for country-specific values and consideration of confounding factoring including dietary habits of population and characteristics of pollutants (Yu et al. 2014) which may vary person to person and region to region, respectively. For example, these kinds of parameters differ significantly in Asian countries from that in European countries, apart from variation in the pollution patterns.

Probabilistic carcinogenic risk assessment for workers exposed to carcinogenic PAHs showed that an ILCR between  $10^{-6}$  and  $10^{-4}$  indicates potential risk, whereas a ILCR greater than  $10^{-4}$  suggests high potential risk (Chen and Liao 2006; NYSDOH et al. 2007). The acceptable level is equal to or lower than  $10^{-6}$ .

Our estimated results suggested that drivers in both sites are exposed to moderate/high carcinogenic risk via both dust ingestion and dermal contact pathways. In fact, cancer risk levels via dermal contact were higher in Lahore (ranging between  $10^{-4}$  and  $10^{-3}$  in AM-L (average  $7 \times 10^{-4}$ ) and between  $10^{-5}$  and  $10^{-3}$  in TP-L (average risk  $3.91 \times 10^{-4}$ ) than in Rawalpindi (ranging  $1.4 \times 10^{-4}$ – $5.6 \times 10^{-4}$  in AM-R (average  $2.4 \times 10^{-4}$ ) and  $7.2 \times 10^{-5}$  to  $4.9 \times 10^{-4}$  in TP-R (average  $2.6 \times 10^{-4}$ ). Similarly, cancer risk levels via ingestion were ranging  $10^{-5}$ – $10^{-4}$  in TP-L,  $1.1 \times 10^{-4}$ – $7.8 \times 10^{-4}$  in AM-L and  $10^{-5}$ – $10^{-4}$  in AM-R and TP-R. The inhalation exposure due to suspended particles was almost

**Table 4** Incremental lifetime cancer risk (ILCR) estimated for the traffic-related cohorts exposed to dust-bound PAHs

Exposure sites			Dermal	Ingestion	Inhalation	$\Sigma$ Risk
Dust samples collected from the traffic police workplace	Rawalpindi	Mean	2.6E-04	1.41E-04	1.09E-08	4.02E-04
		St. Dev	1.43E-04	7.73E-05	5.99E-09	2.20E-04
		Med	1.78E-04	9.59E-05	7.44E-09	2.74E-04
		Min	7.2E-05	3.86E-05	3.00E-09	1.10E-04
		Max	4.9E-04	2.62E-04	2.04E-08	7.48E-04
	Lahore	Mean	3.91E-04	2.11E-04	1.64E-08	6.02E-04
		St. Dev	5.36E-04	2.89E-04	2.24E-08	8.25E-04
		Med	2.72E-04	1.47E-04	1.14E-08	4.19E-04
		Min	7.04E-05	3.80E-05	2.95E-09	1.08E-04
		Max	1.93E-03	1.04E-03	8.09E-08	2.97E-03
Vehicular dust samples	Rawalpindi	Mean	2.4E-04	1.27E-04	1.79E-08	3.62E-04
		St. Dev	1.73E-04	9.34E-05	2.77E-08	2.66E-04
		Med	1.53E-04	8.27E-05	6.41E-09	2.36E-04
		Min	1.4E-04	7.51E-05	5.82E-09	2.14E-04
		Max	5.6E-04	3.03E-04	9.89E-08	8.66E-04
	Lahore	Mean	7.08E-04	3.82E-04	2.96E-08	1.09E-03
		St. Dev	6.15E-04	3.32E-04	2.57E-08	9.47E-04
		Med	2.96E-04	1.60E-04	1.24E-08	4.56E-04
		Min	1.98E-04	1.1E-04	8.30E-09	3.05E-04
		Max	1.45E-03	7.8E-04	6.05E-08	2.23E-03

negligible ( $<10^{-6}$ ) in all samples, and it was much lower than the risk of Beijing traffic police officers (approximately  $4.2 \times 10^{-4}$ – $5.8 \times 10^{-4}$ ) (Liu et al. 2007). The result of cancer risk assessment obtained in this study also raises the concern over the potential effect of a work ambient contaminated with PAHs, and it compels a need for both immediate attention and cleanup/remediation programs at the sites of exposure (Meiners and Yandle 1995) for the health safety of the workers and general public, in particular for that of commuters.

**Conclusions**

This study shows that the work environment may be the most important part of the exposure of traffic policemen and bus drivers to soil-bound PAHs in Lahore and Rawalpindi cities. Exposure of workers to dust, smoke, and other carcinogenic pollutants is highly related to respiratory symptoms and risk of cancer, and for this reason, appropriate health surveillance of workers is necessary. In this study, high levels of PAHs were detected in soil/dust samples from both Rawalpindi and Lahore cities; in particular, the highest PAH concentrations were detected in the samples collected from the public transport of Lahore city. Estimated results of ILCR indicated that traffic policemen and drivers, especially those in the urban area of Lahore, were potentially exposed to high cancer risk via dust ingestion and dermal contact. This study also highlights a need to take serious actions and make strict laws to control air pollution in the big cities of Pakistan and to improve the environmental management and health conditions in workplaces.

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