



Traffic-related lead pollution in roadside soils and plants in Khyber Pakhtunkhwa, Pakistan: implications for human health

I. Ahmad¹ · B. Khan¹ · N. Asad¹ · I. A. Mian² · M. Jamil³

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Abstract

The contamination of lead in roadside soils and in the leaves of selected trees grown on the corresponding roadside land was investigated. Roadside soil and leaf samples of four tree species were collected along the national and provincial highways in five towns of the Khyber Pakhtunkhwa province, Pakistan. The mean lead concentrations in roadside soil samples were 44.8, 39.4, 32.7, 28.4 and 41.7 mg kg⁻¹ for Peshawar, Charsadda, Swabi, Upper Dir and Lower Dir, respectively. For leaves, the mean concentrations were 4.44, 3.48, 5.56 and 5.93 mg kg⁻¹ for *Eucalyptus camaldulensis*, *Dalbergia sissoo*, *Platanus orientalis* and *Morus nigra*, respectively. Human health risks via ingestion, inhalation and dermal contact were estimated numerically in terms of average daily dose, hazard quotient and hazard index for the exposed adults and children. The mean hazard index value for adults was 0.37, and mean hazard index value for children was 3.23. Thus, children were found at high risk of lead-related health issues due to roadside soil pollution.

Keywords Health risk · Lead · Plant contamination · Pollution · Roadside soil · Khyber Pakhtunkhwa

Introduction

The contamination of the natural environment with toxic heavy metals is a global concern. Metal contamination can be a serious problem in developing countries and urbanized regions because of the large number of contaminant sources (Reeder and Shapiro 2003; Cheng and Hu 2010). Among the heavy metals, lead (Pb) is the most abundant in the earth's crust and causes multiple adverse effects to humans and the environment (Cheng and Hu 2010; Markus and McBratney 2001). Lead also is particularly dangerous for children (Shah et al. 2010; Mielke et al. 2010), affecting the development of the brain and nervous system. The Agency for Toxic Substances and Diseases Registry (ATSDR) has classified Pb

as second most toxic heavy metal after arsenic because of its ubiquity and severe toxicity to humans (ATSDR 2015).

Roadside Pb pollution is strongly associated with transportation (Chen et al. 2010; Yan et al. 2018; Fakayode and Olu-owolabi 2003; Mielke et al. 2010). Elevated Pb concentrations were found in roadside soil, plants and vegetation primarily due to automobile exhaust emissions, tire wear, brake pads and lubricants (Yan et al. 2018; Hjortenkrans et al. 2006, 2007; Kluge and Wessolek 2012; Nabulo et al. 2006; Khan et al. 2011b).

Beginning in the 1920s, tetraethyl Pb was added to gasoline at concentrations up to 2000 Pb mg L⁻¹ (Parekh et al. 2002). After discovering the widespread contamination and resulting health issues caused by automobiles burning “ethylated” gasoline (Nriagu 1990), a global movement began to eliminate the practice of adding Pb to gasoline. Japan was the first country to ban the practice in 1986, the USA joined the list in 1996, and Pakistan eliminated Pb as a gasoline additive in 2001. The current US regulatory limit for gasoline is 26 mg Pb L⁻¹; however, typical Pb concentrations in gasoline are < 1 mg L⁻¹ (USEPA 1989a; LEAD Group 2011; Chrastny et al. 2018), whereas in Pakistan a regulatory limit of 0.02 g L⁻¹ Pb⁻¹ is currently followed by the gasoline refineries (Parekh et al. 2002; Paul et al. 2003).

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✉ I. Ahmad
iqbal601@uop.edu.pk

¹ Department of Environmental Sciences, University of Peshawar, Peshawar 25120, Pakistan

² Department of Soil and Environmental Sciences, University of Agriculture, Peshawar 25120, Pakistan

³ Department of Botany, University of Sargodha, Sargodha 40100, Pakistan

Considering that the gasoline marketed in Pakistan had some of the highest Pb contents worldwide and that Pakistan was one of the last countries to phase out leaded gasoline, a survey of Pb in roadside soils and plants is important to evaluate the legacy of leaded gasoline and assess its potential impact on human health. The present study was conducted during August and September 2016 to: (1) measure Pb concentration in roadside soils and plants in five selected towns of Khyber Pakhtunkhwa (KP) province Pakistan; (2) evaluate the health risks of Pb due to ingestion, inhalation and dermal contact pathways for the exposed population.

(KPBOS) reported the total number of registered vehicles as 46,368 in Peshawar, 11,186 in Charsadda, 17,446 in Swabbi, 376 in Upper Dir and 23,241 in Lower Dir (KPBOS 2012). However, a large number of unregistered vehicles also access the roads in these towns and that number increases with increasing population. According to the 2017 census in Pakistan, a total population density of 4.269, 1.616, 1.624, 0.946 and 1.436 million was recorded for Peshawar, Charsadda, Swabi, Upper Dir and Lower Dir districts, respectively.

Materials and methods

Study area

This study was conducted along the national highways such as N-5 (Peshawar), N-45 (Upper Dir and Lower Dir) and provincial highway S-1 (Charsadda and Swabi) (Fig. 1). The traffic in these towns is comprised of light and heavy vehicles such as buses, trucks, cars, rickshaws and motorbikes that consume petrol, diesel and compressed natural gas as fuel. The Khyber Pakhtunkhwa Bureau of Statistics

Soil sampling, preparation and analyses

The soil sampling was carried out within 20 m of the edge of the highways (both sides) and at 0 to 25 cm depth. Samples were collected from those locations along the highways which has no nearby anthropogenic contamination sources such as drain, industrial discharges, waste water irrigation and solid waste dumping sites. A total of 23 roadside soil samples were collected from August 2016 to September 2016 (Fig. 1). A 20-m tape was laid parallel to the highway (on both sides) at each arbitrary site, and 6 subsamples (1 kg each) were collected at equal distances along the tape with a stainless trowel. The subsamples were mixed to form a composite sample representative of that site. Reference soil

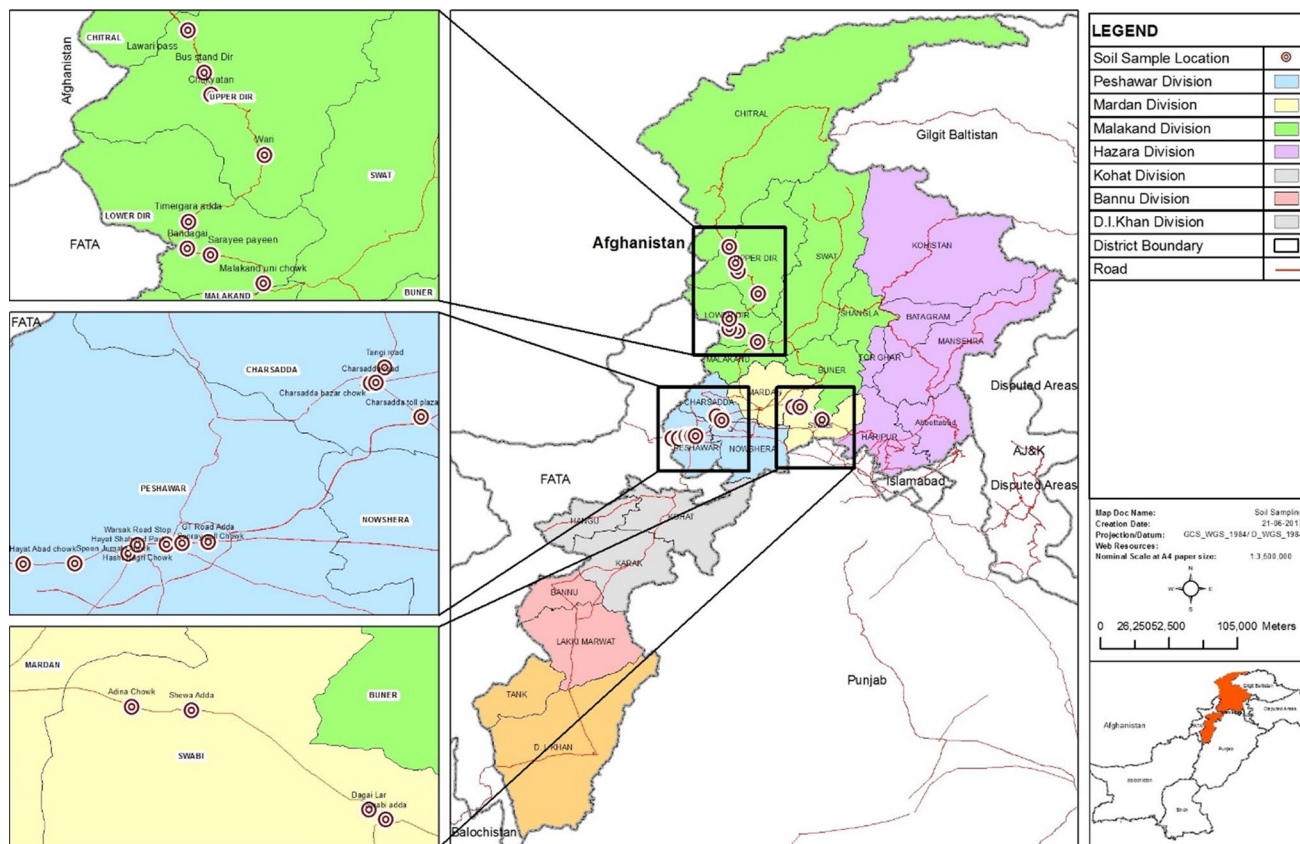


Fig. 1 Location map of the study area showing sampling points

samples (agricultural soil samples with no apparent anthropogenic contamination) were also collected from locations far away from the highways. The soil samples were air-dried, ground and passed through a 2-mm mesh. Basic physico-chemical properties of the roadside soils [i.e., pH, electrical conductivity (EC), organic matter (OM) and texture] were determined (Khan et al. 2016a, b).

For total Pb in soil, 1 g air-dried soil sample was taken into digestion tube placed in digestion block, added 10 mL aqua regia (HCl and HNO₃ at 3:1 v/v) and allowed to stand overnight. The digestion tubes were heated at 150 to 160 °C for 8 to 10 h until digestion. Finally, 3 mL of HClO₄ was added to complete the digestion (Khan et al. 2016b). The extracts were diluted and filtered into centrifuge tubes. All the samples were prepared in triplicates and analyzed for Pb using atomic absorption spectroscopy (AAS, Perkin Elmer 700).

Plant leaves sampling, preparation and analyses

The selected plant species include four species of tree: Red gum (*Eucalyptus camaldulensis*), Indian rosewood (*Dalbergia sissoo*), Oriental plane (*Platanus orientalis*) and Black mulberry (*Morus nigra*), and these tree species were chosen since they are common along the national and provincial highways in the study area, however, distributed randomly. Fully developed leaves (at about 2 m height) were taken from all sides of the crown to give a good average composition at the site. The leaves were cut with stainless scissors from trees within the same distance from the highway as used for soil sampling. A total of 87 leaf samples were collected in paper bags. The dendrometric parameters of the trees such as total height (m) and diameter at breast height (DBH; cm) (1.3 m above the ground) were determined using a measuring tape.

In laboratory, the leaf samples were washed with deionized water, grounded to powder form and oven-dried (60 °C ± 5 °C) for 48 h. For total Pb in leaves, 1 g leaf sample was weighed into the digestion tubes placed in digestion block, added 5 mL of analytical grade nitric acid (HNO₃) and allowed to stand overnight. The samples were heated at 120–130 °C for about 5–6 h until complete digestion and disappearance of brown fumes. The extracts were filtered into centrifuge tubes. All the samples were prepared in triplicates and analyzed for Pb using atomic absorption spectroscopy (AAS, Perkin Elmer 700).

Health risk assessment

Exposure assessment

Health risk assessment involves exposure assessment and risk assessment, while typical Pb exposure pathways for

humans are food chain contamination, direct ingestion, inhalation and dermal contact of the contaminated soils (Fan and Wang 2017; Wu et al. 2015; Khan et al. 2016b). For exposure assessment, we calculated the average daily dose (ADD) levels for the exposed adults and children using Eqs. 1, 2 and 3 as adopted from USEPA (1989a, b, 2001, 2004).

$$ADD_{\text{ingest}} = \frac{C_s \times \text{IngR} \times \text{CF} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (1)$$

$$ADD_{\text{inhal}} = \frac{C_s \times \text{PM} \times \text{InhR} \times \text{EF} \times \text{ET} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (2)$$

$$ADD_{\text{dermal}} = \frac{C_s \times \text{SA} \times \text{CF} \times \text{AF} \times \text{ABS} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (3)$$

$$ADD_{\text{total}} = ADD_{\text{ingest}} + ADD_{\text{inhal}} + ADD_{\text{dermal}} \quad (4)$$

where ADD_{ingest} , ADD_{inhal} and ADD_{dermal} are the average daily dose intakes of Pb from soil ingestion, inhalation and dermal contact pathways ($\text{mg kg}^{-1} \text{ day}^{-1}$), respectively. The ADD_{total} is the sum of all the Pb exposure routes to the body. Description and values of different parameters used in Eqs. 1, 2 and 3 are provided in Table 1.

Risk assessment

The ADD values were used to assess the risk [Hazard Quotient (HQ) and Hazard Index (HI)]. The HQ is measured as the ratio of the ADD to the corresponding reference dose (RfD) or toxicity threshold value, whereas HI is calculated as the sum of HQs for all the exposure pathways. (Poggio et al. 2008; Siciliano et al. 2009). The HQ values were calculated using Eq. 5 adopted from USEPA (1989b, 2004).

$$HQ = \frac{ADD}{\text{RfD}} \quad (5)$$

The RfD values used for oral ingestion, dermal contact and inhalation were 3.50×10^{-3} , 5.25×10^{-4} (Fan and Wang 2017) and $2.00 \times 10^{-4} \text{ mg kg}^{-1} \text{ day}^{-1}$, respectively (USEPA 1989b, 2001, 2004). The overall health risks were assessed using Eq. 6.

$$HI = \sum_{i=1}^n HQ_i \quad (6)$$

An HI value of less than or equal to 1 ($HI \leq 1$) indicates no health risks, whereas an HI value of more than 1 ($HI > 1$) indicates potential of health risks to the exposed population (Man et al. 2010).



Table 1 Values of different variables used for the estimation of human exposure to roadside soils (USEPA 1989b, 2001, 2004, 2011)

Variables	Details	Values
Cs (mg kg ⁻¹)	Concentration of Pb in soil	Measured in this study
IngR (mg d ⁻¹)	Soil ingestion rate	200 for child, 100 for adult
CF (kg mg ⁻¹)	Conversion factor	10 ⁻⁶
EF (d year ⁻¹)	Exposure frequency	350
ED (year)	Exposure duration	6 for child, 24 for adult
BW (kg)	Average body weight	15 for child, 70 for adult
AT (d)	Average time for non-carcinogenic effects	ED×365
SA (cm ²)	Skin surface area available for contact	2800 for child and 5700 for adult
AF (mg cm ⁻²)	Soil to skin adherence factor	0.2 for child, 0.07 for adult
ABS (unitless)	Dermal absorption factor	0.001
InR (m ³ d ⁻¹)	Air inhalation rate for receptor	7.6 for children, 20 for adult
ET (h d ⁻¹)	Exposure time	1
PM (kg m ⁻³)	Particulate (suspended soil) concentration in air	1.36×10 ⁻⁹

Statistical data analysis

Statistical data analysis was carried out using Statistix 10 software. Analysis of variance (ANOVA) was applied to identify the statistical differences. SigmaPlot 10.0 software was used to prepare graphs.

Quality control

Blank reagents as well as standard certified reference soil (GBW07406-GSS-6) and plant (GBW07603-GSV-2) materials were used with each batch of sample preparation and analysis to verify the accuracy and precision of the digestion procedure and analysis. Reference soil and plants were obtained from the National Research Center for Certified Reference Materials in China. The percent recovery for Pb in soil and plant materials was 94% and 92%, respectively. All the apparatus and glassware used were acid washed [5%

HNO₃ (v/v)] and rinsed with deionized water prior to use. The reagents used were of analytical grade.

Results and discussion

Soil and plant parameters

The mean values of different soil parameters are summarized in Table 2. The pH ranged from 7.30 to 8.43, and EC ranged from 0.12 to 0.88 dS m⁻¹. Organic matter content was less than 5% for all the roadside soil samples. The mean sand content was more than 30%, and clay content ≤8.1% for all the soil samples. The environmental fate and transport of heavy metals in soils is strongly influenced by the soil physicochemical parameters (Wilson et al. 2014); sandy soils with low organic matter content generally retain less Pb in the upper soil horizons than soils with high clay and organic matter contents (Reeder and Shapiro 2003).

Table 2 Physicochemical properties of the roadside soils

Location	PH	Electrical conductivity (dS/m)	Organic matter (%)	Texture (%)		
				Sand	Silt	Clay
Peshawar	7.78–8.30*	0.16–0.72	3.40–4.71	31.4–42.8	45.7–53.4	8.10–18.4
	(8.05±0.18)	(0.42±0.24)	(4.21±0.44)	(36.97±3.77)	(48.33±2.67)	(14.7±3.76)
Charsadda	7.82–8.3	0.33–0.88	2.07–4.32	28.6–34.8	53.4–65.0	5.8–14.4
	(8.07±0.20)	(0.58±0.24)	(3.12±0.99)	(31.2±2.87)	(58.7±5.01)	(10.1±3.66)
Swabi	7.84–8.36	0.12–0.44	2.86–4.21	28.9–34.5	54.3–63.4	5.8–13.1
	(8.11±0.23)	(0.27±0.15)	(3.65±0.57)	(31.7±2.63)	(58.8±4.28)	(9.46±3.30)
Upper Dir	7.30–7.98	0.15–0.47	2.10–2.70	51.8–70.8	19.4–40.7	7.5–9.8
	(7.55±0.30)	(0.35±0.14)	(2.50±0.28)	(63.2±8.18)	(27.8±9.24)	(8.95±1.10)
Lower Dir	7.88–8.43	0.20–0.45	2.33–4.10	58.4–71.4	21.0–35.0	6.64–11.9
	(8.16±0.23)	(0.32±0.12)	(2.82±0.85)	(65.0±6.15)	(26.6±6.12)	(8.51±2.32)

*Range (mean ± SD)

Among the trees, all but *E. camaldulensis* are deciduous. The mean height observed for *E. camaldulensis*, *D. sissoo*, *P. orientalis* and *M. nigra* was 18.24 ± 6.81 , 16.48 ± 6.76 , 19.11 ± 7.37 and 9.27 ± 3.27 m, respectively. Likewise, the mean DBH observed for *E. camaldulensis*, *D. sissoo*, *P. orientalis* and *M. nigra* was 22.52 ± 7.27 , 26.44 ± 11.13 , 37.71 ± 12.52 and 25.27 ± 5.16 cm, respectively. Generally, the DBH increased with total height of the tree.

Concentration of Pb in soil and plants

The concentration of Pb in roadside soils and plants is presented in Fig. 2. The range and mean Pb concentration in soil samples was 36.7 to 51.6 (44.8) mg kg^{-1} in Peshawar, 34.2 to 46.2 (39.4) in Charsadda, 25.4 to 47.7 (32.7) in Swabi, 19.0 to 39.8 (28.4) in Upper Dir and 35.5 to 51.3 (41.7) mg kg^{-1} in Lower Dir. These concentrations generally follow the same trends as the number of registered vehicles in each town. The geological background levels of Pb in reference soil samples ranged from 13.3 to 23.5 mg kg^{-1} with a mean concentration of 23.5, 18.1, 13.3, 21.7 and 22.3 mg kg^{-1} for Peshawar, Charsadda, Swabi, Upper Dir and Lower Dir, respectively.

Pearson's correlation analysis indicated that Pb concentration in roadside soil is significantly positively correlated with the number of registered vehicles and total population in each town (Table 3). Similarly, the mean lead concentrations in roadside soils were significantly ($p < 0.003$) higher than reference soils, suggesting that the roadside Pb is mainly contributed by the traffic.

The order of Pb contamination for the different towns was Peshawar > Lower > Dir > Charsadda > Swabi > Upper Dir. The variation in Pb concentration can be attributed to the variable traffic density and regional differences such as wind speed and local terrain along the major roads (Zhang

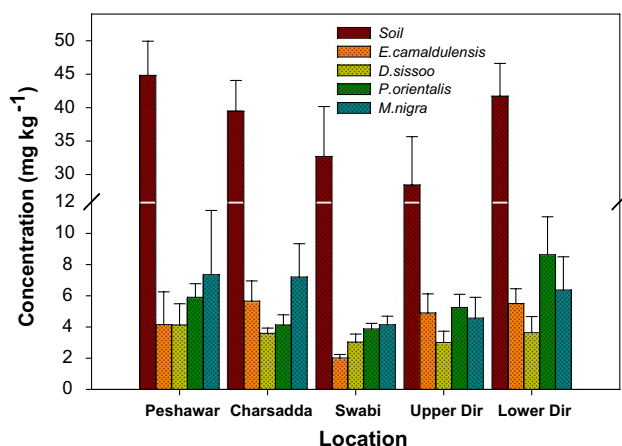


Fig. 2 Mean Pb concentrations in roadside soils and plant leaves in the study area

Table 3 Pearson's correlation (r) among the number of registered vehicles, population and roadside Pb concentration

(r)	Registered vehicles	Roadside Pb
Population	0.92***	0.71*
Roadside Pb	0.83**	1

*, ** and ***significant at p value < 0.05 , < 0.01 and < 0.001

et al. 2015). Previously, Khan et al. (2011a) found Pb concentration along the national highway (N-5) in Pakistan in range of 12.30 to 176.05 mg kg^{-1} with a mean concentration of 36.45 mg kg^{-1} .

The uptake and bioaccumulation of Pb by plants is studied previously (Brown et al. 2016; Hettiarachchi and Pierzynski 2004), and leaf samples of some plant species were found as good indicators of Pb pollution (Tomašević et al. 2013; Safari et al. 2018). The Pb concentrations in plant leaf samples had ranges and mean concentrations of 1.7 to 7.5 (4.44), 2.0 to 6.9 (3.48), 3.1 to 12.3 (5.56) and 2.4 to 14.5 (5.93) mg kg^{-1} , for *E. camaldulenses*, *D. sisso*, *P. orientalis* and *M. nigra*, respectively. The order of contamination was *M. nigra* > *E. camaldulenses* > *D. sisso* > *P. orientalis*. The mean Pb concentrations were significantly ($p < 0.01$) different among the plants. The variation in Pb concentration among the plant species of the same area may be the result of differences in leaf morphology controlling the retention of atmospheric deposition and different plant species having different uptake mechanisms (Blanus et al. 2015).

Health risk assessment

Exposure assessment

The chronic daily exposure estimates are summarized in Table 4. According to USEPA (2011), an ADD value of 1.0×10^{-6} is considered as the health target risk. Health risks are negligible for an ADD value of lower than 1.0×10^{-6} , while an ADD value higher than 1.0×10^{-4} is considered above the permissible guideline limits of the international regulatory agencies (Guney et al. 2010).

The ADD values showed that the exposure dose of Pb is higher for children than adults by approximately a factor of 10. Among the different pathways, oral ingestion of the contaminated soil is projected to be the major exposure route for the population followed by dermal contact and inhalation. This order of exposure is in accordance with the previous studies (Fan and Wang 2017; Hu et al. 2016). Thus, specific attention should be given to address the exposure risk especially for children due to ingestion pathway.

Table 4 Estimated average daily dose values for the exposure pathways

Location	Adults				Children			
	ADD _{ingest}	ADD _{inhal}	ADD _{dermal}	ADD	ADD _{ingest}	ADD _{inhal}	ADD _{dermal}	ADD
Peshawar	6.1×10^{-5}	1.67×10^{-8}	2.5×10^{-7}	6.2×10^{-5}	5.4×10^{-4}	2.8×10^{-8}	1.5×10^{-6}	5.4×10^{-4}
Charsadda	5.4×10^{-5}	1.5×10^{-8}	2.2×10^{-7}	5.4×10^{-5}	4.7×10^{-4}	2.4×10^{-8}	1.3×10^{-6}	4.7×10^{-4}
Swabi	4.5×10^{-5}	1.2×10^{-8}	1.8×10^{-7}	4.5×10^{-5}	3.9×10^{-4}	2.0×10^{-8}	1.1×10^{-6}	3.9×10^{-4}
Upper Dir	3.9×10^{-5}	1.1×10^{-8}	1.6×10^{-7}	3.9×10^{-5}	3.4×10^{-4}	1.8×10^{-8}	9.5×10^{-7}	3.4×10^{-4}
Lower Dir	5.7×10^{-5}	1.6×10^{-8}	2.3×10^{-7}	5.7×10^{-5}	5.0×10^{-4}	2.6×10^{-8}	1.4×10^{-6}	5.0×10^{-4}
Mean	5.1×10^{-5}	1.4×10^{-8}	2.0×10^{-7}	5.1×10^{-5}	4.5×10^{-4}	2.3×10^{-8}	1.3×10^{-6}	4.5×10^{-4}

Table 5 Estimated health risk values for the exposed population

Location	Adults				Children			
	HQ _{ingest}	HQ _{inhal}	HQ _{dermal}	HI	HQ _{ingest}	HQ _{inhal}	HQ _{dermal}	HI
Peshawar	0.02	0.31	0.12	0.44	0.15	2.69	1.03	3.87
Charsadda	0.02	0.27	0.10	0.39	0.14	2.37	0.90	3.41
Swabi	0.01	0.22	0.09	0.32	0.11	1.96	0.75	2.83
Upper Dir	0.01	0.20	0.07	0.28	0.10	1.71	0.65	2.46
Lower Dir	0.02	0.29	0.11	0.41	0.14	2.51	0.95	3.60
Mean	0.01	0.26	0.10	0.37	0.13	2.25	0.86	3.23

Risk assessment

The health risk values for adults and children are presented in Table 5. The estimated HQ and HI values were highest for Peshawar followed by Lower Dir, Charsadda, Swabi and Upper Dir. The mean HI values were 0.37 for adults and 3.23 for children (Table 4). The exposed children have a high probability of health risks ($HI > 1$). This is due to the behavioral and physiological characteristics of children, including hand-to-mouth activities in soil and higher respiration rates per unit body weight. (Fan and Wang 2017; Karim and Qureshi 2014). The general assumption is that orally ingested Pb is fully absorbed by the gastrointestinal tract (Luo et al. 2012).

Conclusion

Contamination of Pb is prevalent in the roadside soils and plants along the national and provincial highways in Khyber Pakhtunkhwa. The roadside Pb contamination was in the order of Peshawar > Lower Dir > Charsadda > Swabi > Upper Dir. Among the different plants, highest mean Pb concentrations were found for *M. nigra* followed by *P. orientalis*, *E. camaldulenses* and *D. sissoo*. Oral ingestion of the Pb contaminated soil is the major exposure route for the exposed adults and children followed by dermal absorption of the soil

particulates adhered to the skin and direct inhalation. The exposed population especially children are at high risk of Pb-related health effects. Measures should be taken to remediate and protect the roadside soils from Pb contamination.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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