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Vehicular mediated emissions of polycyclic aromatic hydrocarbons in roadside soils of Shanghai

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This study evaluates the impact of vehicular emissions on polycyclic aromatic hydrocarbons (PAHs) in roadside soils in Shanghai during the COVID-19 lockdown period. Soil samples from roadside lawns were collected, with PAH concentrations ranging from 153 to 5639 ng g^{-1}. A significant reduction in PAH levels compared to their pre-COVID-19 levels (Kruskal–Wallis H test, $p < 0.05$) was observed in surface soil samples, highlighting the contribution of traffic and coal combustion to urban pollution. Source identification, using molecular diagnostic ratios and principal component analysis, revealed that vehicular emissions were the primary contributors to PAHs in Shanghai's roadside soils. The toxic equivalent quantity for benzo[a]pyrene concentrations in the soil samples was associated with these sources. The incremental lifetime cancer risk model indicated that adult exposure to PAHs in the soil posed health risks greater than 10^{-6} but lower than 10^{-4}, suggesting a low-risk level. These findings suggest that targeted measures in the transportation sector could improve urban soil quality and reduce associated health risks.

Keywords Polycyclic aromatic hydrocarbons, COVID-19 pandemic, Roadside lawns soils, Vehicular-mediated emissions, Health risk assessment, Source identification

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic compounds that consist of two or more fused aromatic rings. They are primarily generated by the incomplete combustion of organic materials such as coal, petroleum, and biomass[1]. There are several types of PAHs, ranging from two-ring compounds like naphthalene to more complex structures such as benzo[a]pyrene. Due to their chemical stability and lipophilic nature, PAHs are persistent in the environment, accumulating in various media including soil, water, and air, and often persisting for long periods[2]. They are widely recognized as environmental pollutants with significant health risks. Prolonged exposure to PAHs, even at low levels, can lead to serious health issues, including mutagenic and carcinogenic effects, due to their tendency to bioaccumulate and transfer through the food chain[3,4].

Human activities are the predominant source of PAHs in the environment, especially in urbanized and industrialized areas. Major contributors include vehicle emissions, industrial processes, and fossil fuel combustion, with vehicular emissions accounting for a significant share of PAH pollution in urban settings[5–7]. Although natural events such as volcanic eruptions and forest fires can also release PAHs, their impact is relatively small compared to anthropogenic sources[8]. The growing reliance on motor vehicles in densely populated urban areas has exacerbated PAH emissions, making traffic emissions a primary target for pollution control strategies[9,10]. Numerous studies have demonstrated the link between vehicular emissions and elevated levels of PAHs in urban soils and air, with PAHs from exhaust and tire wear particles accumulating in the surrounding environment, particularly in roadside soils[11–14].

Despite extensive research on the spatial distribution, transport, sources, degradation, and health impacts[15], their high persistence in soils poses, poses challenges for remediation. PAHs can remain in soils, sediments, and water for extended durations, and natural degradation processes are often insufficient for their complete removal[16,17]. Moreover, end-of-pipe treatments for PAHs are both costly and inefficient, prompting a shift toward source reduction as a more sustainable approach[18]. This strategy is particularly relevant in the transportation and industrial sectors, where emissions are directly linked to PAH contamination[10]. Despite the increasing emphasis on source reduction, ongoing fossil fuel use means that PAH emissions persist, and there is a need for more focused research on emission control at the source. Evaluating source reduction measures is essential

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for providing scientific insights that can inform environmental management policies and urban sustainability practices.

The COVID-19 lockdown provided a unique opportunity to study the impact of reduced human activity on environmental pollution[19–21]. During the lockdown, transportation and industrial activities in China decreased significantly, leading to a substantial reduction in motor vehicle emissions[22,23]. This created a natural experimental condition for studying the short-term effects of reduced human activity on PAH concentrations in urban soils. The distribution of PAHs in urban soils is closely related to human activities, particularly in highly industrialized and densely populated cities such as Shanghai[24]. Each year, Shanghai consumes large amounts of coal, gasoline, and other petroleum products, leading to the long-term accumulation of PAHs in the city's soils, posing threats to environmental quality and human health[20].

This study uniquely leverages the COVID-19 lockdown as a natural experiment to evaluate the immediate effects of reduced vehicular emissions on PAH concentrations in urban soils—a research gap not adequately addressed in previous studies. The findings aim to support the development of sustainable transportation regulations and pollution mitigation strategies.

Materials and methods

Sample collection and preparation

A total of 72 surface soil samples (0–1 cm) were collected in October 2021 from roadside lawns along major roads to ensure adequate spatial coverage and statistical representation of different road types and traffic densities in Shanghai, including Huancheng Road, Zhonghuan Road, Yueluo Road, Jiasong Road, and Shenzhuan Road (Fig. 1). At each site, multiple subsamples were collected and combined into form a composite sample. During sampling, the climatic conditions were characterized by mild temperatures (approximately 15–20 °C) and low precipitation, factors that may influence pollutant deposition. After collection, samples were sealed in Teflon bags, wrapped in aluminum foil, and stored in iceboxes at 4 °C during transportation to the laboratory. The samples were freeze-dried, ground into a fine powder[25]. During the sieving process, precautions such as using a pre-washed 80-mesh sieve were taken to minimize particle loss. However, any potential loss of fine particles is acknowledged as a limitation in our study.

Sample extraction and analysis

PAHs were extracted using an accelerated solvent extraction system (Dionex ASE 350, Thermo Fisher Scientific, Waltham, MA, USA) and analyzed on a GC-MS system equipped with an Agilent 5975 C mass spectrometer (Agilent Technologies Inc., USA) and an HP-5 capillary column (30 m × 0.25 mm, 0.25 μm film thickness), operated in selective ion monitoring (SIM) mode. The instrument accuracy was verified through routine calibration with certified standards[25]. Approximately 5 g of soil were mixed with six deuterated PAH surrogate standards (naphthalene-d_8, phenanthrene-d_{10}, acenaphthene-d_{10}, fluorene-d_{10}, pyrene-d_{10}, and benz[a] anthracene-d_{12}) to monitor recovery rates. The samples were subjected to accelerated solvent extraction using dichloromethane. Extracts were purified using Florisil solid-phase extraction cartridges (1.0 g, 6.0 mL, Angel Laboratory Technologies, Shanghai, China). Elution was performed with 8 mL of a dichloromethane-hexane (1:1, v/v) mixture, followed by nitrogen evaporation to approximately 0.4 mL. The final extract was transferred to a brown injection vial for analysis. Sixteen priority PAHs, identified by the US Environmental Protection Agency (US EPA), were analyzed using gas chromatography-mass spectrometry (GC-MS). The temperature program began as an isothermal 60 °C for one minute, then ramped linearly from 60 °C to 150 °C at a rate of 10 °C min^{-1}, held at 150 °C for five minutes, increased from 150 °C to 200 °C at a rate of 5 °C min^{-1}, held at 200 °C for five minutes, ramped from 200 °C to 250 °C at a rate of 5 °C min^{-1}, held for five minutes, and finally ramped from 250 °C to 300 °C at a rate of 5 °C min^{-1}. Finally, the program was held at 300 °C for 5 min. Chrysene-d_{12} was used as an internal standard to quantify the PAH concentrations.

Quality assurance and quality control (QA/QC)

For every 20 samples, procedural, transportation, and field blanks were analyzed to check for contamination. PAH quantification was performed using calibration curves generated from certified standards, ensuring reliable and reproducible measurements. Although the recovery rates for surrogate standards ranged from 57.1 to 94.4%, the potential loss of fine particles during the sieving process is acknowledged as a limitation. The recovery for individual surrogates was as follows: naphthalene-d_8 (57.1 ± 11.4%), acenaphthene-d_{10} (76.8 ± 8.8%), phenanthrene-d_{10} (93.0 ± 8.6%), fluorene-d_{10} (92.9 ± 5.9%), pyrene-d_{10} (94.4 ± 5.9%), and benzo[a] anthracene-d_{12} (90.8 ± 3.8%).

Source identification and toxicity apportion with a PMF model

Source identification

Numerous techniques for identifying sources have been documented in recent years. Molecular diagnostic ratios[1,26], stable carbon isotope method of monomer hydrocarbon[17], radiocarbon isotope method of monomer hydrocarbon, principal component analysis (PCA), and positive matrix factorization (PMF) are commonly used to distinguish between different sources[27]. These models allow for a comprehensive analysis of the contributions from various activities, including the combustion of coal, biomass, and petroleum[28]. PCA was used to identify data clusters and underlying patterns, and PMF provided a quantitative estimate of source contributions, thereby complementing each other in the source identification process[29]. Therefore, the PMF model, PCA, and molecular diagnostic ratios [Flt/(Flt + Pyr) and IcdP/(IcdP + BghiP)] were used in this study to identify the likely origins of PAHs in the samples. The Flt/(Flt + Pyr) ratios less than 0.4 are linked to petrogenic sources; ratios between 0.4 and 0.5 indicate the burning of coal combustion; and ratios greater than 0.5 indicate the vehicular emission[30]. Furthermore, IcdP/(IcdP + BghiP) ratios less than 0.2 are linked to petrogenic sources; ratios between 0.2 and 0.5

indicate the traffic emission; and ratios greater than 0.5 indicate the coal and biomass combustion[31]. The PMF software uses EPA PMF 5.0 provided by the US EPA. The model parameters, including the number of factors, uncertainty estimates, and diagnostic statistics (Q value, residuals), are detailed in Supplementary material.

Toxicity apportion

The PMF model was also used to apportion toxicity based on Benzo[a]pyrene toxic equivalents (BaP_{TEQ})[32]. The BaP_{TEQ} values were calculated using the PAH concentrations and their respective toxic equivalent factors (TEFs)[33,34]. This approach allowed for the assessment of the carcinogenic potential of the detected PAHs from different sources. A full list of TEFs used in the calculations is provided in Table S1.

Incremental lifetime cancer risk (ILCR)

To evaluate the potential health risks of PAHs, the ILCR model, developed by the US EPA, was employed. The model considered three exposure pathways: ingestion, dermal absorption, and inhalation of soil particles. Exposure data and parameters used for the ILCR calculations were obtained from the US EPA's guidelines[34] and are detailed in Table S2.

Results and discussion

Concentrations of PAHs in roadside lawns soil samples

When comparing the pre-lockdown and post-lockdown periods, a significant reduction in PAH concentrations were observed during the lockdown. The average total PAH concentration across all samples was 1199 ng g^{-1}, with a range of 153–5639 ng g^{-1} (see Table 1 and Table S3). In addition, the PAHs composition in the soil samples of both sub-regions was dominated by moderate molecular weight PAHs (4 ring PAHs) and high-molecular-weight (HMW) PAHs (5 ring PAHs and 6 ring PAHs) (Fig. S1). As demonstrated by Table 1, the concentrations of PAHs in this study were comparatively lower than their pre-COVID-19 levels when compared to similar roadside lawn areas in Shanghai, such as industrial regions and areas near highways (1776 ng g^{-1})[35–38] and green spaces like parks, greenbelts, and woodland areas (1888 ng g^{-1})[37–43]. Additionally, a Kruskal-Wallis H test was conducted on the data from this study and previous studies to examine whether there were differences between the groups. The analysis results show that, since the p-value ($p = 0.00053$) is less than 0.05, there is a significant difference between the two groups of data (see Fig. S2). This decrease is attributed to the drastic reduction in road traffic during the lockdown, which significantly lowered emissions from vehicular sources. Similar reductions in PAH levels during lockdowns have been reported in other cities[44–46].

The observed decrease aligns with global trends observed during the COVID-19 lockdown, where transportation emissions saw marked reductions due to stringent lockdown measures. However, the relatively short duration of the lockdown may limit the overall impact on long-term soil PAH accumulation, particularly for more persistent PAH compounds.

Source identification of PAHs

The potential sources of PAHs in the soil samples of Shanghai were identified through the use of the PCA and molecular diagnostic ratios. The molecular diagnostic ratio cross plots are illustrated in Table 2 and Fig. S3. The IcdP/(IcdP + BghiP) ratio in most soil samples collected from roadside lawns in Shanghai ranged from 0.2 to 0.5, indicating that the PAHs originated from traffic emissions. Additionally, the Flt/(Flt + Pyr) ratio, which fell between 0.5 and 0.7, pointed to vehicular emissions as a significant source of PAHs[25]. Overall, the diagnostic ratio results of soil samples collected from roadside lawns in Shanghai indicated that incomplete fuel combustion emissions from vehicular emission are the primary source of PAHs.

By examining the loading of the particular PAHs found in the soil samples, two main components (PC1 and PC2) were found, accounting for 88.6% of the total variations (see Table 2). PC1 displayed a significant positive loading of NaP, Ace, Flu, Phe, Ant, Flt, Pyr, BaA, Chr, BbF, BkF, BaP, IcdP, DBahA, and BghiP. This component clarified 79.4% of the total discrepancies. PC2 accounted for 9.2% of the variance, mainly consisting of NaP, Acy, Flu, and Ant, with additional loadings of Ace, Phe, Flt, Pyr, BaA, Chr, BkF, and BaP. Analysis of the soil samples suggested a combination of combustion sources from vehicle emissions as well as coal combustion, based on the outcomes of the principal components. The PCA revealed that PAH emissions from vehicle emissions and coal combustion were common sources in the soil samples. These results from the PCA align with those obtained

Soil types	Range (ng g^{-1})	Mean (ng g^{-1})	Sampling time (year)	References
Industrial land, urban parks, university campuses, and residential areas soils	1562–7017	3279	2007	35
Urban and industrial areas soil	62–31900	1700	2007	37
Central urban areas soils	83–7220	1970	2011	38
Urban (Pudong) and suburban soils	19–6320	807	2011	39
Surface soils from the Yangtze River Delta	10–3059	267	2014	41
Agricultural soils from industrial sites	258–535	360	2014	42
Road green spaces, parks, and woodland green spaces soils	Not detection – 18,916	1888	2016	43
Roadside lawns soils	163–14289	1776	2019	36
Roadside lawns soil samples	153–5639	1199	2021	This study

Table 1. Summary of PAH concentrations in soil samples in Shanghai.

PAHs	PC1	PC2
NaP (2-rings)	0.35	0.69
Acy (3-rings)	− 0.1	0.86
Ace (3-rings)	0.88	0.26
Flu (4-rings)	0.66	0.67
Phe (3-rings)	0.89	0.39
Ant (3-rings)	0.6	0.63
Flt (3-rings)	0.91	0.34
Pyr (4-rings)	0.9	0.35
BaA (4-rings)	0.97	0.23
Chr (4-rings)	0.89	0.35
BbF (5-rings)	0.96	0.15
BkF (5-rings)	0.95	0.22
BaP (5-rings)	0.9	0.38
IcdP (6-rings)	0.94	0.1
DBahA (5-rings)	0.96	0.19
BghiP (6-rings)	0.94	0.14
Eigenvalues	12.7	1.5
Variance %	79.4	9.2
Cumulative variance %	79.4	88.6
Identified sources	Coal combustion, and vehicular emission	Petrogenic and coal combustion

Table 2. The total variance explained and component matrix of PAHs in roadside lawn soil samples in Shanghai.

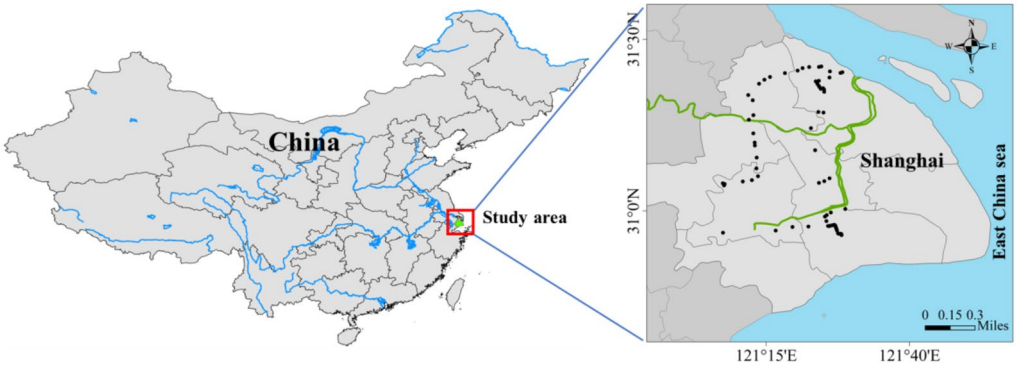

Fig. 1. Sampling stations in Shanghai.

using the molecular diagnostic ratios method. The findings are consistent with previous studies[37,40], indicating that vehicular emissions are one of the dominant sources of PAHs in urban soils. The significant reduction in PAH levels during the lockdown reinforces the potential of traffic control measures in mitigating environmental pollution, thereby offering new insights into the short-term impacts of emission reductions (Fig. 1).

Toxicity apportion with a PMF-TEQ method

The PMF-TEQ technique is utilized to determine the varying carcinogenic potentials of different sources of PAHs emissions. Initially, the PMF model was employed to analyze a range of components, from three to six, to pinpoint the most significant factors influencing the data[47]. Subsequently, the four-factor model was selected for further investigation as it was deemed most suitable for the two distinct soil types under study (see Table S4). In this investigation, the PMF model identified four primary sources of PAH emissions: coal burning, biomass burning, vehicle emissions, and petrogenic sources. Additionally, the source patterns of each PMF component for PAHs in different soil types are illustrated in Fig. 2.

The results suggested that in soil samples, factor 1 accounted for 2.3% of the total measured PAHs, primarily containing NaP, Flu, and Ace. The prevalence of low molecular weight PAHs indicated a petroleum release. Factor 2 was responsible for 56.0% of the total PAHs, dominated by Flt, Pyr, BaA, Chr, BbF, BkF, and BghiP, which are associated with vehicle exhaust. Factor 3 accounted for 29.3% of the total PAHs, with high loadings on Ant, Flu, Pyr, BbF, and BkF, known as tracers of coal combustion. Factor 4 included prominent compounds such

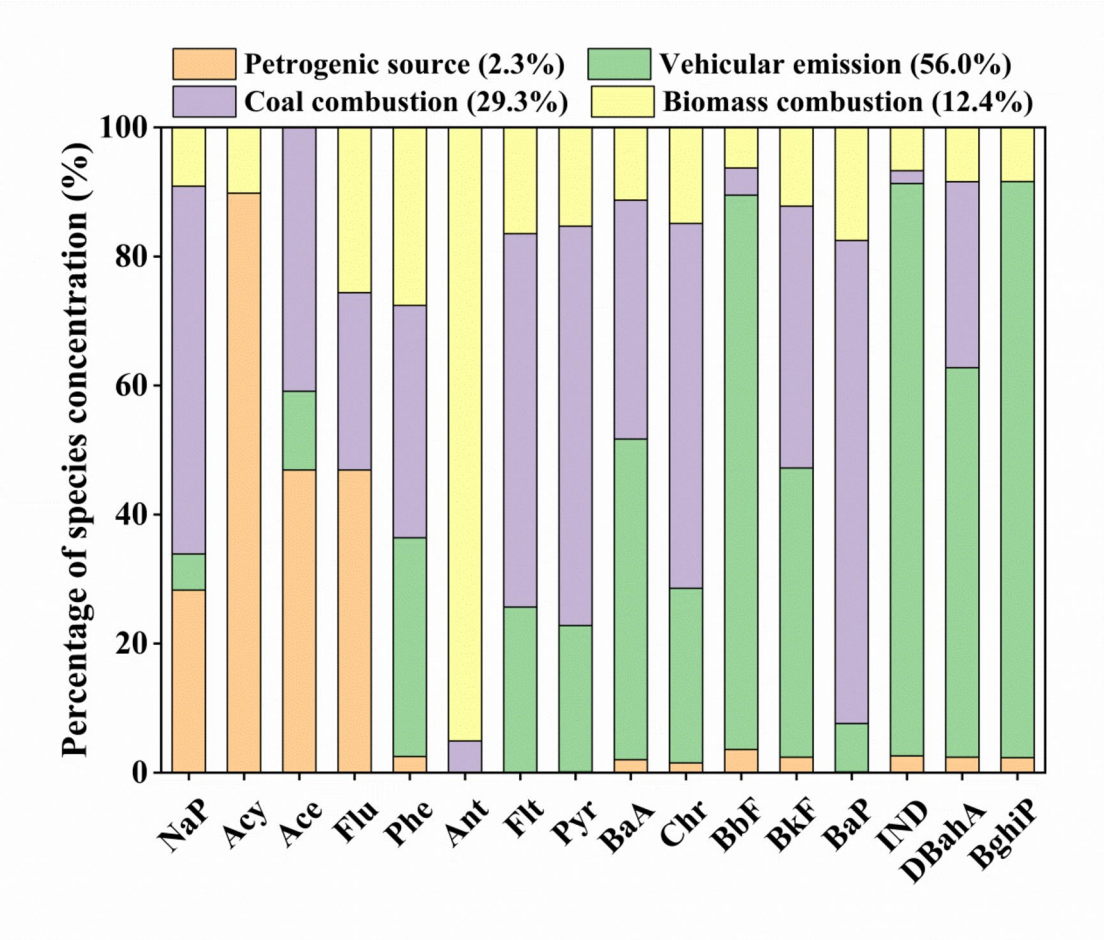

Fig. 2. Source profiles of each PMF factor for PAH in roadside lawns soil samples in Shanghai.

as BkF, BaP, IcdP, DBahA, and BghiP, identified as significant markers of biomass combustion. Therefore, Factor 4 likely represents biomass burning and explains 12.4% of the total PAHs.

The BaP_{TEQ} was also evaluated based on the TEF values of the 16 PAHs species in order to determine their level of toxicity in the soil samples from roadside lawns. Accordingly, the BaP_{TEQ} for each of the 16 PAH compounds in soil samples from roadside lawns was, with a range of 10.1–6075.8 ng g^{-1}, 202.4 ng g^{-1} (median values from the lognormal distribution). The highest estimations in risk prediction were typically derived from the 95th percentile BaP_{TEQ} concentrations. Therefore, the soil samples had a greater concentration (4104.8 ng g^{-1}) of BaP_{TEQ}.

The 95th percentile BaP_{TEQ} concentrations for the 16 PAHs chemicals in both sub-regions are shown by the source contributions in Fig. 3. Carcinogenic risks from vehicle emissions (54.8%, 2249.4 ng g^{-1}) and coal combustion (30.1%, 1235.5 ng g^{-1}) were highest in soil samples; these were followed by carcinogenic risks from biomass combustion (12.4%, 508.9 ng g^{-1}) and petrogenic sources (2.8%, 114.9 ng g^{-1}). Therefore, the main causes of the BaP_{TEQ} concentrations for PAHs in the soil samples were vehicle emissions and coal burning.

Incremental lifetime cancer risks for adults exposed to PAHs in soils

A quantitative evaluation was carried out to assess the ILCRs faced by residents of Shanghai who are exposed to soil PAHs through various means of exposure[24]. As indicated in Fig. 4, the estimated 95th percentile of ILCR values for individuals exposed to soil PAHs in soil samples were 2.2×10^{-5}. The US EPA has put forward a categorization system that classifies cancer risks into three categories: safe-level ($ILCR < 10^{-6}$), low-risk ($10^{-6} < ILCR < 10^{-4}$), and high-risk ($ILCR > 10^{-4}$)[19,26]. Adults exposed to PAHs in either type of soil had an ILCR falling within the range of higher than 10^{-6} but lower than 10^{-4}, suggesting a threshold of low risk.

Conclusions

In conclusion, this study demonstrated that vehicular emission reductions during the COVID-19 lockdown led to a significant decrease in PAH concentrations in Shanghai's roadside soils. Source apportionment confirmed that vehicular emissions were the dominant contributor. These findings provide strong scientific evidence for implementing stricter vehicular emission controls and urban pollution mitigation policies.

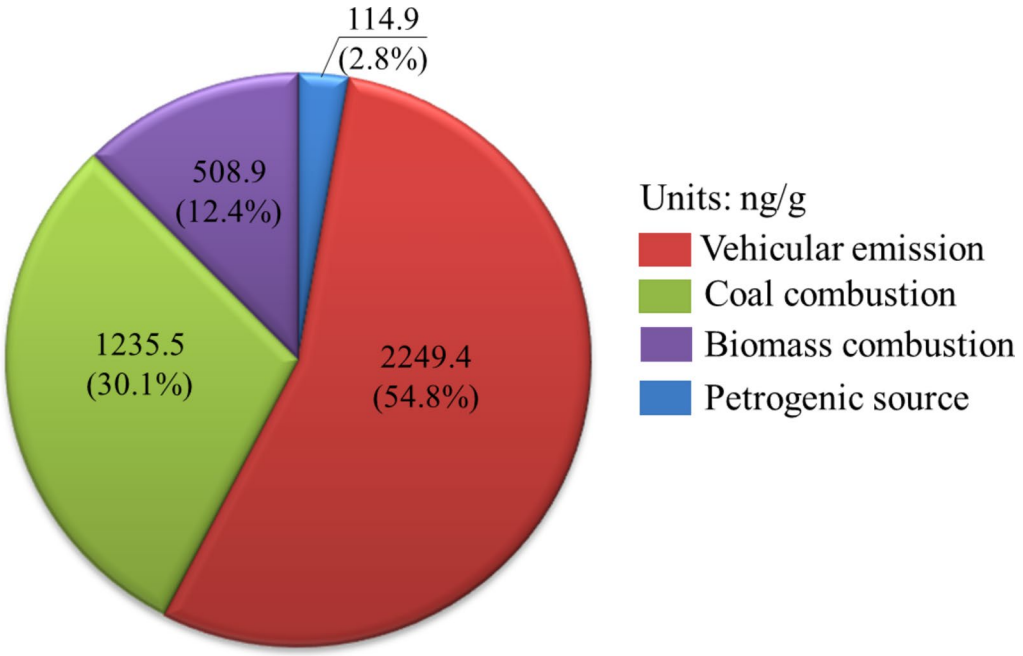

Fig. 3. Source contributions to the 95th percentile BaP_{TEQ} concentrations for the 16 PAH compounds in roadside lawn soil samples.

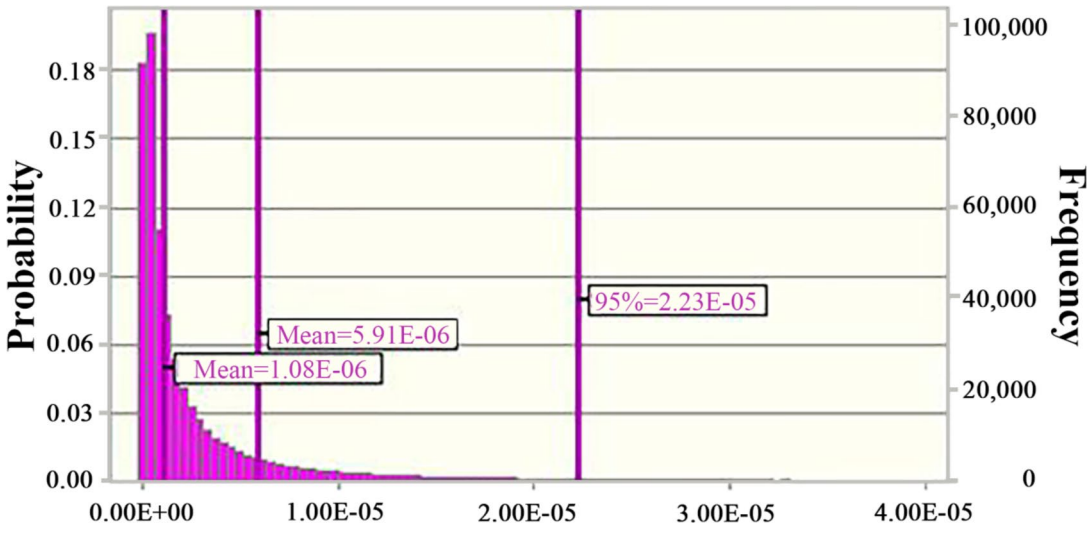

Fig. 4. Predicted probability density functions of ILCR for adults exposed to the PAH in roadside lawn soil samples.

However, limitations include the short sampling period and lack of comprehensive pre-lockdown data. Future research should focus on long-term monitoring and improved pollution control strategies. These findings offer valuable guidance for policymakers to develop sustainable urban planning strategies that mitigate vehicular emissions and improve environmental health.

Data availability

The data and materials that support the findings of this study are available from the corresponding author on reasonable request.

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Declarations

Ethics approval and consent to participate

This study does not contain any studies involving animal or human participants performed by any of the authors. The manuscript in part or in full has not been submitted as a preprint server published anywhere before submitting it to Environmental Science and Pollution Research. All authors were participated in this work.

Competing interests

The authors declare no competing interests.

Consent for publication

All authors agree to publish.

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