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Impact of PAHs compounds on air quality in Maragheh city: Probabilistic risk assessment and source apportionment

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ABSTRACT

This study explores the concentrations and spatial distribution of polycyclic aromatic hydrocarbons (PAHs) in the ambient air of Maragheh city, Iran, while evaluating their potential health implications. PAHs levels were examined in PAHs-bound to particulate matter samples collected from diverse locations across the city. The results showed that in all sampling points, there was contamination by PAHs. The mean total PAHs concentration was 11.5 $ng.m^{-3}$, with Benzo[*a*]pyrene (BaP) emerging as the predominant compound. Comparative analysis with other cities revealed relatively lower BaP levels in Maragheh, yet surpassing WHO guidelines in 92 % of samples. Spatial assessment heightened pollution in areas characterized by heavy traffic and industrial operations. Based on PCA analysis, it appears that 74 % of PAHs compounds originate from vehicle emissions, 13 % from the combustion of petroleum, and 6 % from a possible petroleum source. Health risk appraisal uncovered escalated carcinogenic and mutagenic hazards, especially among children. While risks remained below USEPA thresholds, ongoing monitoring and targeted interventions are advised to mitigate PAHs pollution in Maragheh and similar urban locales. Future endeavors should prioritize source elucidation, health impact assessments, and public awareness initiatives to safeguard community well-being.

1. Introduction

In ambient air polycyclic aromatic hydrocarbons (PAHs) boast a complex chemical structure, comprising carbon, hydrogen, nitrogen, sulfur, or cyclopentane rings. Notable examples include naphthalene, anthracene, phenanthrene, pyrene, fluoranthene, benzo(a)-pyrene, benzo(e)pyrene, perylene, benzo(g, *h*, i)perylene, and coronene. These compounds exhibit remarkable resistance to combustion, enabling them to persist in the environment for extended periods. Additionally, certain PAHs possess the ability to volatilize into the atmosphere. They are broadly categorized into two groups based on their ring composition, with varying molecular weights and boiling points [1,2]. The generation of PAHs arises from both natural events, like forest fires, and human activities, encompassing industrial operations, transportation, and cooking practices. Particularly, forest fires stand out as a significant natural contributor to PAHs [3].

The International Agency for Research on Cancer (IARC) has classified specific PAHs, like benzo[*a*]pyrene, as Group 1 carcinogens, underscoring their potential health risks. Advanced analytical techniques have facilitated the identification of numerous airborne particles with carcinogenic properties, including several PAHs [4,5].

Various challenges persist in estimating the quantity of these pollutants, as highlighted by studies investigating tire wear emissions [6]. The literature on PAHs in ambient air emphasizes their persistence as environmental pollutants globally. Studies, including those by Okuda et al. [7] in Chinese cites, Zhang et al. [8] in Kanazawa, Japan, and

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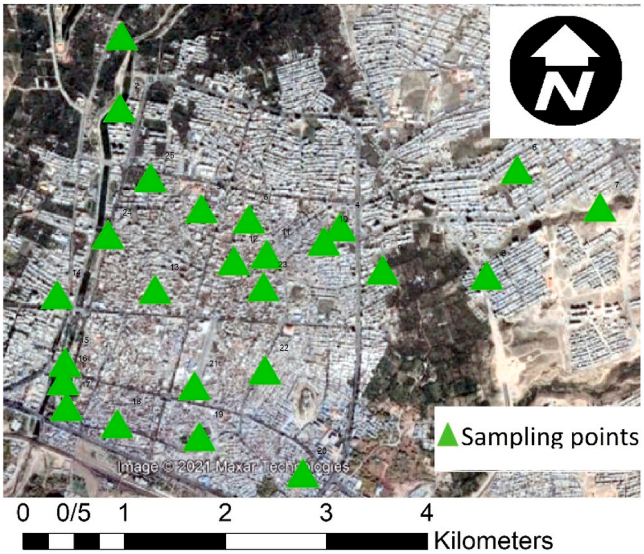

Fig. 1. The sampling points of the study area and its surroundings.

Oleagoitia et al. [9] in Basque Country, Spain, have extensively analyzed PAHs concentrations in the PM2.5 fraction of air in urban industrial areas [7–9]. They identified vehicular emissions as the primary source, with concentrations ranging from 0.85 to 9.86 ng/m^{-3}. In Sabzevar, Iran, a 2021 study assessed PAHs concentrations in PM and their sources, revealing significant contributions from petrogenic sources, with PM-bound PAHs levels ranging from 1056.2 to 848.5 ppb [10]. Similarly, research in Tehran from 2018 to 2019 investigated spatial differentiation in PM2.5-bound PAHs characteristics, indicating higher concentrations in high-traffic roadside areas, especially during cold seasons [11]. Correlation analysis implicated the combustion of liquid fossil fuels as the dominant source, with a concerning incremental lifetime cancer risk (LTCR) via inhaling PM2.5-bound PAHs [12].

Atmospheric concentrations of PAHs exhibit a wide range, influenced by emission sources, chemical reactivity, and meteorological factors. Despite concentrations peaking during winter months, recent studies have indicated a decreasing trend in atmospheric PAHs levels over time [13,14]. Guidelines established by the World Health Organization (WHO) underscore the health risks associated with exposure to benzo[*a*]pyrene, prompting regulatory measures in several nations [15–17]. Given the scarcity of evidence concerning pollutant sources in urban settings, particularly regarding PAHs in ambient air in developing countries, our study aims to evaluate ambient PAHs concentrations bound to particulate matter, conduct source apportionment analysis, and assess carcinogenic risks associated with inhalation exposure to PAHs.

2. Materials and methods

2.1. Study site

The research area is located within Maragheh city, nestled in the northwest region of Iran and situated within East Azerbaijan province. Maragheh boasts a population of approximately 180,000 residents inhabiting an area spanning 30 square kilometers which 55 percent reported as adult and 45 percent children. It holds the distinction of being the fourth largest urban center in northwest Iran and the second largest city within East Azerbaijan province as a touristic city. Renowned for its historical significance, Maragheh stands as one of Iran's ten ancient cities and is recognized as the astronomy capital of the nation [18].

The climate of Maragheh, in terms of long-term temperature and monthly precipitation (over 30 years or more), falls into the Cs class according to the Köppen-Geiger climate classification. This classification indicates moderate climatic conditions with a dry summer [19].

2.2. Sampling procedure

In this study, twenty-five sampling points were selected at various locations throughout the city of Maragheh (Fig. 1). These points were chosen to cover the entire urban area as high and low traffic. The sampling device employed was a suspended particulate matter sampler acquired from Zist-Sepehr Beyhagh company. This sampling device was equipped with a polycarbonate filter with a diameter of 12 millimeters, housed between two protective plates to collect PM particles with less than 50 microns while shielding the filter from wind and rain [20]. In each sampling points, the sampler was installed at a height of approximately 2 m above ground level. The sampling duration for all of stations was 3 months (from May 22, 2022, to August 21, 2022). The concentration of PM-PAHs was calculated in ng/m^3 using the guideline method proposed by Wagner and Leith, as well as the Castillo et al. [21,22], method, after detecting the values.

This study examined 15 PAHs, which are: Naphthalene (NapH), Acenaphthylene (Ac), Acenaphthene (Ace), Fluorene (Fl), Phenanthrene (Phen), Anthracene (Anth), Fluoranthene (F), Pyrene (Py), Benzo[*a*]anthracene (BaA), Chrysene (Chr), Benzo[*b*]fluoranthene (BbF), Benzo[*a*]pyrene (BaP), Indeno[1,2,3-cd]pyrene (IcdPy), Dibenzo[*a*,*h*]anthracene (DbahA), Benzo[*g*,*h*,*i*]perylene (BghiP).

2.3. Sample extraction and PAHs identification

The extraction protocol followed the method outlined in the studies by Arfaeinia and Aslam [23,24]. Initially, the filters were kept in dichloromethane for 20 h. Subsequently, ultrasonication (BANDELIN, GMMINIZO, Germany) was performed for 15 min to facilitate extraction, followed by a 5-min resting period (repeated three times). Following preparation and extraction, the detected values of PAHs compounds were determined using a GC-MS. The specifications of the GC/MS instrument and the relevant protocol are presented in Table S1.

2.4. Quality control and quality assurance (QA/QC)

Instrument calibration was conducted on a daily basis using calibration standards. Each detection batch involved repetitions of detections, spiked samples, and control samples. The limit of detection (LOD) for PAHs varied from 0.0002 to 0.0004 ng/m^3, with the LOD determined using 10 control samples. Inter-assay accuracy assessment relied on the recovery percentage (82–103 %) and desorption efficiency. PAHs calibration curves were created using pure standards (from Sigma-Aldrich) across concentrations ranging from 10 to 1000 μg/m^3.

2.5. Health risk assessment

Assessment of the carcinogenic risk resulting from inhaling a blend of PAHs compounds was performed by determining the concentration of benzo[*a*]pyrene equivalent (B[*a*]Peq) through deterministic modeling. Eq. (1) was applied to derive the B[*a*]Peq equivalent. BEC represents the benzo[*a*]pyrene equivalent of atmospheric PAHs.

$$BEC = \Sigma Ci \times TEFi \tag{1}$$

Ci: PAHs value, TEF: toxicity equivalency factor for the PAHs compound.

Details of each are provided in Table S2.

Eq. (2) was used to extract the daily inhalation exposure (DIE).

Ti: daily exposure time to PAHs, BECi: benzo[*a*]pyrene equivalent of PAHs, IR: breath rate (m^3/day).

$$DIF = \Sigma BECi \times IR \times Ti \tag{2}$$

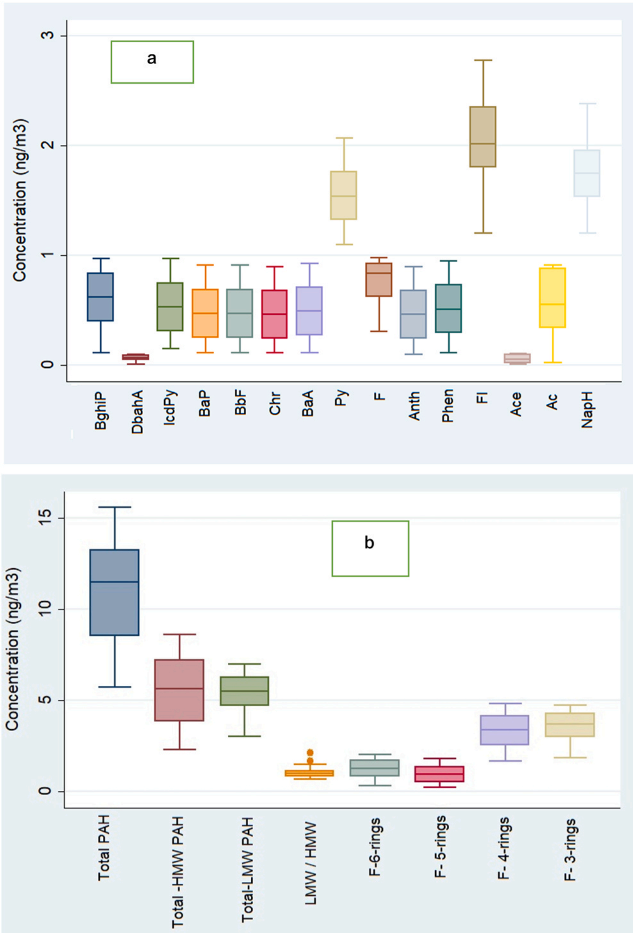

Fig. 2. Descriptive statistic of ambient PAHs congeners in Maragheh, a) level of each \PAHs compounds, b) summarize of cumulative level of a different class of PAHs compounds.

Eq. (3) was employed to determine the lifetime cancer risk (LTCR) from breath exposure, with details of the factors in the equation provided in the Table S2 [25].

$$LTCR = \frac{SF \times DIE \times EF \times ED \times CF}{BW \times AT} \quad (3)$$

Monte Carlo simulation method was employed for stochastic health risk modeling. To achieve this, the Crystal Ball software (version 11.1.2.4, Oracle, Inc., USA) was utilized, capable of conducting 1000 interactions. Stochastic sensitivity analysis was also performed to assess how different factors rank in their impact on the risk associated with LTCR.

2.6. Spatial analysis

Utilizing ArcGIS10.1, a graphical stochastic model was developed for unsampled sampling points, allowing the overlay of pollution layers to generate a contamination classification map. This map visually represents darker shades for polluted points and areas shaded in green and brown indicating heightened pollution levels [37–39]. To analyze the spatial distribution of PAHs, the kriging method in GIS software was employed to categorize air PAHs concentrations across Maragheh.

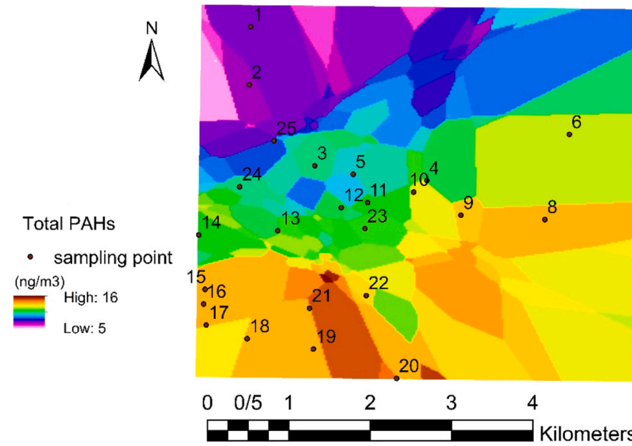

Fig. 3. Spatial distribution of the PAHs congeners in Maragheh city.

3. Result and discussion

3.1. PAHs levels in Maragheh city

The findings from assessing the concentrations of PAHs compounds in Fig. 2 are represented in the corresponding box plots. All samples yielded values surpassing the detection limit of the device, indicating air pollution and its origins. The highest and lowest total PAHs levels were 5 and 15 ng.m^{-3}, respectively, with an average of 11.5 ng.m^{-3} calculated for total PAHs. The concentration range of BaP, considered the most significant compound, spanned from 0.1 to 0.9 ng.m^{-3}, with a median of 0.47 ng.m^{-3}. These figures fall within the recommended range set by the European Union, which suggests staying below 1 ng.m^{-3} annually [26]. The WHO has proposed a threshold of 0.12 ng.m^{-3} for BaP, a limit exceeded by 92 % of the samples in our study. The corresponding concentrations for lifetime exposure to B[*a*]P producing excess lifetime cancer risks of 1/10,000, 1/100,000 and 1/1,000,000 are approximately 1.2, 0.12 and 0.012 ng/m^3, respectively [27].

In Tehran, to measure PAHs bound to PM, PAHs levels of 16 compounds ranged from 56.98 ± 15.91 to 110.35 ± 57.31 ng.m^{-3} in summer and from 125.87 ± 79.02 to 171.25 ± 73.94 ng.m^{-3} in winter were measured. they results indicates that air temperature and solar radiation intensity are influential in reducing PAHs levels. Additionally, BaP concentrations ranged from 2.1 to 8.3 ng.m^{-3}, which exceed those reported in the present study and WHO guidelines. Tehran experiences high traffic volume [28].

In the coastal city of Bushehr, located in southern Iran, PAHs and BaP concentrations were recorded as 0.56 and 18 ng.m^{-3}, respectively, which is similar to the present study. The similarity in emission sources and relatively similar geographical extent could be the reason for this resemblance with our study [29].

In Naples, Italy, PAHs and BaP concentrations were reported as 1.1 and 2.0 ng.m^{-3}, respectively, across four seasons, while another study in the city of Bursa, Turkey, detected PAHs concentrations at 2.05 ng.m^{-3} [30,31]. These studies demonstrate the presence of PAHs levels in respiratory air worldwide.

In the present study the highest levels were observed in Fl, Naph, and Py, with median concentrations of 2, 1.7, and 1.5 ng/m^3, respectively, as shown in Fig. 2 box plots. Additionally, median levels of total low molecular weight PAHs (LMW-PAHs) and total high molecular weight PAHs (HMW-PAHs) were 5.6 and 5.5 ng/m^3, respectively, indicating a uniform distribution of these substances. However, previous research in Sabzevar, Iran, reported LMW levels three times higher than HMW-PAHs in airborne particles [10], whereas another study in Rafsanjan, Iran, found HMW-PAHs to be five times higher than LMW in roadside soil [32]. The primary reason for this disparity remains unclear, possibly due to larger particles settling faster or natural atmospheric processes

Table 1
PAHs ratios used with their typically reported values for specific processes emissions [42,43].

PAHs ratio	Value range	Probable emission source	This study Mean (Standard deviation)
IcdPy/ (IcdPy + BghiP)	< 0.2	Petrogenic	0.48 (0.06)
	0.2–0.5	Petroleum combustion	
	> 0.5	Grass, wood, coal combustion	
(BaA/BaA + Chr)	0.2–0.35	Coal combustion	051 (0.02)
	> 0.35	Vehicular emissions	

like biological and photochemical breakdown [33,34]. HMW compounds are generally more resistant to degradation and more toxic than LMW [35,36]. Climate conditions and air temperature likely play a significant role in degradation, given the hot, dry climates of Rafsanjan and Sabzevar compared to the moderate climate of Maragheh. Consequently, faster degradation of LMW in warmer areas could lead to lower levels compared to HMW-PAHs [10,32].

3.2. spatial distribution of PAHs compounds

As Fig. 3 northern area, depicted in blue, corresponds to Maragheh's orchards with low PAHs levels. Conversely, the central portion of the map, shown in blue, indicates the polluted old urban fabric. The southwestern, southeastern, and southern regions, highlighted in brown, exhibit significant pollution, likely due to the high traffic volume of entry and exit roads, including a transit route. This distribution map of PAHs compounds was similar to a study that investigated BTEX spatial and temporal trends in Maragheh. The reason for the pollution in the southern part of the Maragheh city is attributed to the presence of fuel pump stations, transit roads, transportation of diesel trucks and buses, the train station, and heavy traffic in the vicinity [19].

3.3. Sources identification of PAHs emission

Regarding PAHs compounds, like BTEX compounds including benzene, toluene, ethylbenzene, and xylene, the relative proportions of these compounds to each other can indicate potential emissions. Because the proportion of these compounds varies across different sources and even within [40,41]. In this study, the ratios of PAHs compounds are presented in Table 1. We utilized two ratios, IcdP/(IcdP + BghiP) and (BA / BaP + Chr), and identified two potential emission sources for this study based on past research: petroleum combustion and vehicular emissions [42,43]. Maragheh is primarily an agricultural city with no nearby industries, vehicle traffic and gas station activities can be explained by interpreting PAHs ratios. Behnami et al.'s study on air BTEX in Maragheh supports this interpretation [19]. These findings are consistent with a study conducted in Spain and the capital of Iran, Tehran, as the use of solid fuels in Iranian cities has been phased out [9,

Table 2
Factor patterns of the three principal components.

	Component		
	PC1	PC2	PC3
BghiP	**0.913**	0.268	-0.027
DbahA	**0.674**	0.505	0.016
IcdPy	**0.987**	0.118	-0.104
BaP	**0.971**	0.191	-0.097
BbF	**0.970**	0.207	-0.100
Chr	**0.979**	0.171	-0.102
BaA	**0.962**	0.223	-0.096
Py	**0.981**	-0.032	-0.110
F	**0.890**	0.099	0.132
Anth	**0.977**	0.144	-0.102
Phen	**0.981**	0.162	-0.102
Fl	0.316	**0.884**	-0.067
Ace	-0.120	**0.936**	0.020
Ac	-0.112	-0.038	**0.990**
NapH	0.682	**0.682**	-0.091
Proportion of variance (%)	74	13	6.7
Cumulative %	74	87	93. 8
Eigenvalue	0.769	0.145	0.079
Probable Source	Vehicular emission	Petroleum combustion	Petroleum

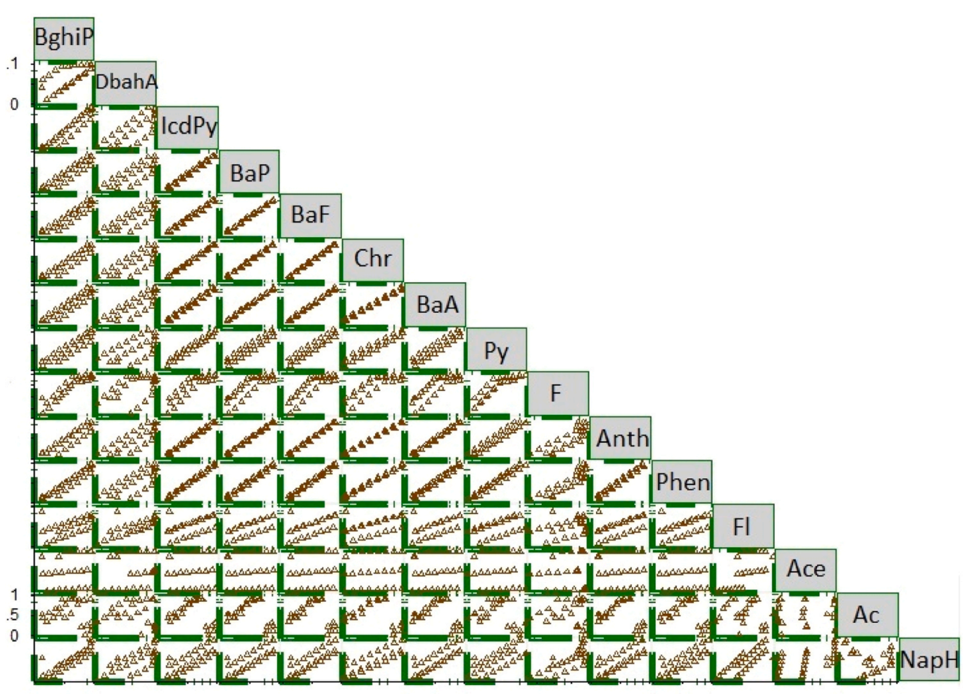

Fig. 4. Spearman correlation coefficients of PAHs congeners.

Table 3
Lifetime cancer risk from inhaling ambient PAHs.

	Population group							
	Children		Adolescents		Adults		Seniors	
	Boy	Girl	Male	Female	Male	Female	Male	Female
DIE (ng/day)								
mean	9.0	8.8	13.1	12.5	19.1	14.3	12.0	10.9
S.D[a]	± 4.1	± 4.0	± 5.9	± 5.7	± 8.7	± 6.5	± 5.4	± 4.9
LTCR								
mean	2.09E-07	2.15E-07	1.55E-07	1.57E-07	1.72E-07	1.51E-07	1.16E-07	1.01E-07
S.D	± 9.68E-08	± 9.96E-08	± 7.21E-08	± 7.30E-08	± 7.96E-08	± 6.99E-08	± 5.39E-08	± 5.0E-08

[a] S.D: Standard deviation.

44].

In studies focusing on environmental contaminants, which are grouped homogeneously, statistical correlation is utilized to explore statistical significance. Spearman's correlation among PAHs compounds is depicted in Fig. 4. Within this research, Ace and Ac demonstrated notably disruptive correlations with other compounds. The correlations of DbahA, F, and NepH with PAHs compounds were not particularly robust, yet they exhibited strong correlations with other compounds. These findings are similarly observed in the investigation of Sabzevar city in Iran [10]. The commonality in emissions could stem from the predominant contribution of traffic and pollution from gasoline-driven vehicles, although some of these findings are also evident in the research conducted in south of Brazil, Naples city, Italy [10,30,45].

Principal component analysis (PCA) and multivariate statistical analyses are utilized to determine potential sources of PAHs compound emissions in a grouped manner. PCA was conducted using Stata version 14 software to extract components based on PAHs compound values (Table 2). As Table 2 the compounds were categorized into three groups (PCs). According to the results, 74 % of the variables belonged to the first group (PC 1), including Anth, F, Py, BaA, Chr, BbF, BaP, IcdPy, DbahA, Fl, and BghiP. These compounds seem to originate from vehicular emissions. The second group (PC 2) covers 13 % of the compounds, including NapH, Ace, and Phen. PC2 findings suggest petroleum combustion as a source. In the third group (PC 3), which comprises 6 % of the compounds, Ac was prominent, indicating a potential petroleum source. Similar results have been observed in studies conducted in Northern India and Rafsanjan city in Iran [32,46].

3.4. Health risk assessment

The deterministic pathway's depiction of lifetime cancer risk levels resulting from inhaling ambient PAHs is presented in Table 3. It also includes data on ambient PAHs levels through inhalation (DIE). The findings indicate that children face the highest lifetime cancer risk from inhaling ambient PAHs. While risks are relatively similar across all age groups, they are lower compared to findings from other studies and fall below the threshold level recommended by USEPA (i.e., 1 × 10ˆ-6) [10,28,47,48]. Fig. S1 displays the results of risk assessment using the stochastic approach. Modeled results show that the highest 95th percentile of LTCR values is for children (3.3 × 10ˆ-7), and the lowest is for seniors (1.8 × 10ˆ-7), both below the USEPA threshold. In previous studies in Iranian cities, due to high air pollution, these values have been higher and reported significantly. Therefore, children are categorized as sensitive groups, with pollutant concentration being identified as the most influential factor in sensitivity analysis regarding potential cancer risks [49].

Suggestions for future research include regular monitoring of PAHs levels, identifying pollution sources, studying the long-term health effects on vulnerable populations, implementing emission reduction strategies, and raising public awareness about PAHs risks

4. Conclusion

The assessment of PAHs levels in Maragheh revealed significant air pollution, with total PAHs levels averaging 11.5 ng/m^3 and BaP levels exceeding WHO thresholds in 92 % of samples. Pollution varied across the city, with higher levels in central urban areas due to traffic. Major sources of PAHs pollution were identified as petroleum combustion and vehicular emissions. Children were found to have the highest lifetime cancer risk from PAHs exposure. Recommendations include regular monitoring, identifying pollution sources, studying health impacts, implementing emission reduction strategies, and raising public awareness.

CRediT authorship contribution statement

Ali Soleimani: Investigation, Formal analysis. **Zahra Atafar:** Writing – original draft, Validation, Software. **Sepideh Nemati-Mnsor:** Writing – review & editing, Methodology, Investigation. **Morshad Ahmed:** Writing – review & editing, Methodology, Investigation. **Hesam Ahmady-Birgani:** Software, Methodology. **Parisa Ravan:** Writing – review & editing, Investigation. **Mohammad Miri:** Investigation, Data curation, Conceptualization. **Amir Mohammadi:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Amir Mohammadi reports was provided by Maragheh University of Medical Sciences. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Declaration of Competing Interest

The authors declare no conflicts of interest associated with this study.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.toxrep.2024.101686.

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