RESEARCH ARTICLE

Seasonal trend and source identification of polycyclic aromatic hydrocarbons associated with fine particulate matters (PM$_{2.5}$) in Isfahan City, Iran, using diagnostic ratio and PMF model

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Abstract

Particulate matters (PMs) and their associated chemical compounds such as polycyclic aromatic hydrocarbons (PAHs) are important factors to evaluate air pollution and its health impacts particularly in developing countries. Source identification of these compounds can be used for air quality management. The aim of this study was to identify the sources of PM$_{2.5}$-bound PAHs in Isfahan city, a metropolitan and industrialized area in central Iran. The PM$_{2.5}$ samples were collected at 50 sites during 1 year. Source identification and apportionment of particle-bound PAHs were carried out using diagnostic ratios (DRs) of PAHs and positive matrix factorization (PMF) model. The results showed that the concentrations of PM$_{2.5}$ ranged from 8 to 291 µg/m^3 with an average of 60.2 ± 53.9 µg/m^3, whereas the sum of concentrations of the 19 PAH compounds (ΣPAHs) ranged from 0.3 to 61.4 ng/m^3 with an average of 4.65 ± 8.54 ng/m^3. The PAH compounds showed their highest and lowest concentrations occurred in cold and warm seasons, respectively. The mean concentration of benzo[a]pyrene (1.357 ng m^{-3}) in December–January, when inversion occured, was higher than the Iranian national standard value showing the risk of exposure to PM$_{2.5}$-bound PAHs. Applying DRs suggested that the sources of the PAHs were mainly from fuel combustion. The main sources identified by the PMF model were gasoline combustion (23.8 to 33.1%) followed by diesel combustion (20.6 to 24.8%), natural gas combustion (9.5 to 28.4%), evaporative-uncombusted (9.5 to 23.0%), industrial activities (8.4 to 13.5%), and unknown sources (2.8 to 15.7%). It is concluded that transportation, industrial activities, and combustion of natural gas (both in residential-commercial and industrial sectors) as the main sources of PAHs in PM$_{2.5}$ should be managed in the metropolitan area, particularly in cold seasons.

Keywords Air pollution · Positive matrix factorization model · Organic pollutants · Metropolitan area

Introduction

Air pollution is one of the world's largest health and environmental problems. It includes a mixture of gases and solid or liquid particles suspended in the atmosphere. Among major air pollutants, particulate matters (PMs) are important as

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they significantly affect the human health, atmospheric chemistry, and global climate change (Li et al. 2020; Zong et al. 2016). PMs are classified by their size which smaller ones cause most health problems (Amil et al. 2016). Several studies have showed that PMs, especially PM$_{2.5}$ (i.e., the particles with aerodynamic diameter smaller than 2.5 µm) can adversely affect human health (Ouyang et al. 2020; Pope et al. 2011). PMs contain various organic pollution such as hydrocarbons and inorganic pollutants such as heavy metals, whereas their chemical composition depends on the emission sources. Therefore, identification and quantification of PM sources and their associated compounds is necessary to improve strategies for effective air quality management (Waked et al. 2014).

Polycyclic aromatic hydrocarbons (PAHs) are the main group of organic pollutants associated with PMs which

consist of two or more fused benzene rings (Luo et al. 2019; Wu et al. 2014; Hassanvand et al. 2015). PAHs both in gas and particle phase can emitted to the environment from natural processes such as incomplete combustion of organic matters, volcanic eruptions, and forest fires (Wang et al. 2008). However, in urban and industrial areas, these compounds are formed mainly through incomplete combustion of fossil fuels (Luo et al. 2019; Qin et al. 2015; Huang et al. 2015; Mostert et al. 2010; Lai et al. 2017). Due to adverse health effects, PAHs have attracted considerable attention by researchers and are considered as priority pollutants (Wang et al. 2020a, 2017). Therefore, for efficient control of air pollution caused by PAHs, identification of the sources of these compounds in the atmosphere is very important.

Diagnostic ratios (DRs) are a common tool for identifying the main PAHs sources (petrogenic or pyrogenic sources) in air, water, sediment, and soil samples as well as organism tissues (Tobiszewski and Namiesnik 2012). PAHs with a various range of molecular weight could be originated from pyrolysis process, while the lowest molecular weight compounds are dominated in petrogenic sources (Abdel-Shafy and Mansour 2016; Tobiszewski and Namiesnik 2012).

Multivariate receptor models are useful approach for identification and apportionment of pollutant sources at a receptor site. Among multivariate receptor modeling techniques, positive matrix factorization (PMF) is widely used for source apportionment. The model was developed by Paatero and Tapper (Paatero and Tapper 1994; Paatero 1997) and has been widely used for source apportionment of various environmental pollutants including organic and inorganic compounds (Sun et al. 2020; Li et al. 2020; Vlachou et al. 2019; Men et al. 2018; Karanasiou et al. 2009). One of the main advantages of this model is that the methodology can be applied without the need for data on source emission compositions. Ambient data of chemicals and their uncertainty are required for running the model. Another advantage of the model is to utilize a point-by-point least squares minimization scheme. Such a weighting scheme allows the inclusion of missing values, and noisy and uncertain data in the analysis through giving low weights (Khan et al. 2012).

Isfahan as a main metropolitan city of central Iran exhibits serious air pollution problems during the last three decades because of industrial development and increasing the population. According to the previous studies, PMs are the major air quality problems in Isfahan city (Norouzi and Khademi 2015; Norouzi et al. 2017; Soleimani et al. 2018) which are associated with other pollutants such as heavy metals and organic pollutants originated from anthropogenic and natural sources (Soleimani et al. 2018). To the best of our knowledge, no source apportionment studies have so far been performed to identify sources and quantify their contributions to ambient PMs and particle-bound PAHs in Isfahan city.

According to the previous studies, fine particulate matter (i.e., $PM_{2.5}$) is the main factor affecting air quality of Isfahan city (Soleimani et al. 2018). Providing valuable information regarding to the concentration and sources of air pollutants such as PAHs is useful for air pollution management. It is therefore of outmost importance to investigate the associated pollutants PM-bound PAHs which can adversely affect the health of people living in the region. The main objective of this study was therefore to determine the concentrations, seasonal variations, and sources of PAHs in $PM_{2.5}$ of an industrial metropolitan city in Iran as a developing country. This will do by the use of DRs in combination with PMF model. The results from this study revealing the potential sources of PAHs in particulate matters during cold and warm seasons may be used by managers and policy makers in action plan of air pollution control and management in the region.

Material and methods

Sampling sites

This study was conducted in Isfahan city, the second largest industrial and the third most populated city, in Iran with more than 2 million inhabitants (https://stat.mpo-es.ir). It located in central Iran (51° 39′ 40″ E, 32° 38′30″ N) with an average elevation of 1590 m (a.s.l.) covering an area about 482 km² (Mansouri and Hamidian 2013).

The climate of study area is principally characterized by hot, dry summers and cold winters. The mean annual temperature is 16.2 ℃ with the warmest month in July (high average 29.3 ℃) and the coldest in January (low average 2.9 ℃) (Mansouri and Hamidian 2013). Isfahan is a receptor of air pollution released from different kinds of industries including steel and iron industries, petrochemical plants, oil refineries, cement and brick factories, power plants, and lead and zinc mines, within city and the surroundings. These industries also have a significant effect on the transportation and traffic capacity. Moreover, because of existence an arid area in north east of the city, it subject to frequent dust storm events during the year (Norouzi and Khademi 2015). Therefore, the city has severe air quality degradation due to the high levels of the atmospheric PMs. To have better understanding of the sources of $PM_{2.5}$ in the study area, stratified and judgmental sampling approach was conducted according to guidance of US-EPA (2002) from December 2017 to September 2018 (including 4 seasons) and 200 samples were collected from 50 sites (Fig. 1) coverage of about 158 km² based on population density, road traffic, and distance to industrial zones inside the study area. In addition to industrial plants around the city, several industrial zones inside the
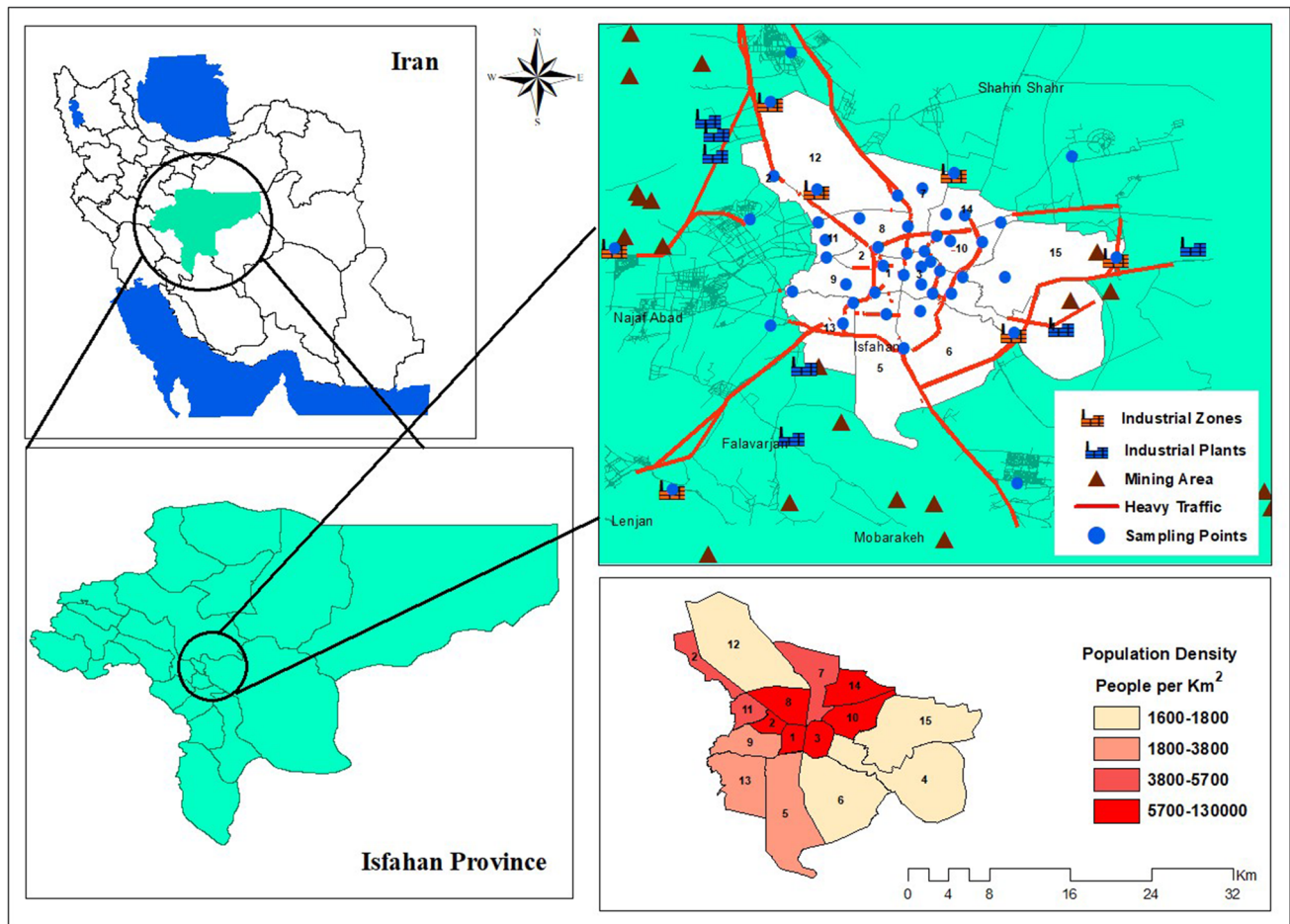
Fig. 1 Location of sampling sites, Isfahan city, Iran

area were also considered in selection of sampling points. It should be noticed that the fall sampling period included the December–January months when the region was faced to the air temperature inversion, while in February–March period which was accounted as winter season, there was not the meteorological phenomenon. According to the European guide on air pollution source apportionment with PMF model, the number of samples should be ≥ 100 (Belis et al., 2014). However, according to the literature (Johnson et al., 2011; Watson et al., 2002), PMF model can also be used for source apportionment if a sufficient number of chemically characterized ambient samples is available (more than 50).

Sampling of $PM_{2.5}$

The samples were collected during 24 h using a high-volume air sampler (TISCH, USA) running at 1.1 m^3 min^{-1}. Quartz filter fibers (with the size of 20.3 cm × 25.4 cm, Sigma-Aldrich) were used for sample collection. All filters were put inside a muffle furnace at 500 °C for 3 h prior to sampling for sure to be clean of any organic compounds. In order

to minimize the influence of water adsorption, the filters were kept in a desiccator for 24 h before and after sampling. $PM_{2.5}$ mass was measured using a 5 decimal digit balance. Then, the samples were stored at -18 °C in a freezer prior to analysis (Khan et al. 2015).

Extraction of PAHs

Each sampled filter was cut into the same size pieces (with area of 3.7993 cm^2) and three of them (determined by primary laboratory tests) including marginal and central parts were selected randomly and used for the extraction using a modified method of EPA-TO-13A (US-EPA, 1999). A sonication system was used for the extraction of PAHs using 5 mL of pentane/dichloromethane 90:10 for 10 min. Before extraction, 4 µL the lab PAH surrogate internal standard mixture (100 times diluted of stock standard) was added to the sample. After extraction, the filter was removed from the extract and 0.5 mL of isooctane was added to the extract and then the solvent volume was reduced to 1 mL. Then 4 µL PAH recovery standard mixture (1000 times diluted of stock

standard) was added to the extract and then it was transferred to a GC vial. The list of internal and recovery standards for the quantification of PAHs is given in Table S1. The blank filters were also used in the field without sampling just to find any contamination of the sample transport to the laboratory. The analyzed PAHs were naphthalene (Nap), acenaphthene (Acy), acenaphthylene (Ace), fluorene (Flu), dibenzothiophene (Dbt), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fl), pyrene (Pyr), benzo(a)anthracene (B[a]A), chrycene (Chr), benzo(b)fluoranthene (B[b]F), benzo(k)fluoranthene (B[k]F), benzo(e)pyrene (B[e]P), benzo(a)pyrene (B[a]P), indeno[1,2,3-cd]pyrene (I[c]P), dibenzo[a,h]anthracene (D[ah]A), and benzo [g,h,i]perylene (B[ghi]P).

Analysis of PAHs

The extracts were analyzed for PAHs and alkyl PAHs by gas chromatography–mass spectrometry using an Agilent Technologies 5975C inert XL MSD with electron ionization operating in selected ion monitoring (SIM) mode. A 60-m HP-5 capillary column with 0.25-µm inner diameter and 0.25-µm film thickness was used. The injection port was kept at 300 °C and 1 µL sample was injected in the splitless mode. The flow rate of helium as carrier gas was 1.1 mL/min. The column temperature was initially held at 40 °C for 2 min, ramped with 25 °C/min to 100 °C, and then programmed at 5 °C/min to 315 °C and held for 14 min. The transfer line, ion source, and quadropole temperatures were 315 °C, 230 °C, and 150 °C, respectively. Values of 55 m/z were monitored in 12 groups with 13 m/z each, with a dwell time of 25 ms, according to Table S2 (Gallotta and Christensen 2012).

MassHunter Quantitative Analysis version B.07.00 was used for quantification of peaks (Agilent technologies, Inc.) and the data was quantified using the internal standards method and corrected for recoveries. Standard calibration curve in the range of 0.003–2 µg/mL was used for each target compound with internal and recovery standards. The overall recovery of the compounds ranged from 71 to 108%. The estimated limit of detections (LOD) for the PAH compounds (ranged from 0.01 to 0.21 ng m^{-3}) are shown in Table S3 which were mostly in the range of previous researches (e.g., Hoseini et al., 2016).

Source apportionment techniques

Diagnostic ratio

In order to identify the source of PAHs, the ratio values of An/(An + Phe), (anthracene to anthracene plus

phenanthrene), Flu/(Flu + Pyr), (fluoranthene to fluoranthene plus pyrene), BaA/(BaA + Chr), (benzo[a]anthracene to Benzo[a]anthracene plus chrysene), IP/(IP + Bghi), (Indeno[1,2,3-cd]pyrene to Indeno[1,2,3-cd]pyrene plus Benzo[ghi]perylene), and low molecular weight to high molecular weight (LMW/HMW) PAHs were widely used by previous researchers (Liang et al. 2019; Suman et al. 2016; Khan et al. 2015; Callén et al. 2011; Katsoyiannis et al. 2007). In this study, the diagnostic ratios were calculated and then compared with the standard values. Table S4 shows the standard values for diagnostic ratios of PAHs.

Positive matrix factorization

In this study, positive matrix factorization (PMF) 5.0 model (US EPA 2014) was applied to quantify the contribution of various PAH sources. Details of the model are described in EPA PMF 5.0 Fundamentals & User Guide (US EPA 2014; Norris et al. 2014). The user manual guideline was closely followed in this study. Briefly, two input files are required by the model: (1) the measured concentrations of the species and (2) the estimated uncertainty of the concentration. The quality of data was assessed based on the signal to noise ratio (S/N) and the percentage of samples above method detection limit (MDL). Those species which had S/N ≥ 2 were classified as strong and those with S/N between 0.2 and 2 and with S/N < 0.2 were considered as weak and bad in quality, respectively. The bad quality values were excluded from further analyses.

The second data file was the uncertainty; for concentrations below and above the MDL, which was estimated by Eqs.(1) and (2), respectively (US EPA 2014).

$$\text{Unc} = \frac{5}{6} \times \text{MDL} \tag{1}$$

$$\text{Unc} = \sqrt{(\text{Error Fraction} \times \text{Concentration})^2 + (0.5 \times \text{MDL})^2} \tag{2}$$

The PAH concentrations and uncertainties were introduced to the model. In principle, the PMF model is a factor analysis algorithm based on the following equation:

$$XIJ = \sum_{K=1}^{P} gikfkj + eij \tag{3}$$

where X_{IJ} is a data matrix (the Jth species in the Ith sample), P is the number of factors, f_{kj} is the source chemical composition (source profile), g_{ik} is the source contributions to each observation, and e_{ij} is the residual matrix that cannot be explained by the model.

The PMF model was used to minimize the object function, Q (E) through an iterative algorithm as shown in Eq. (4).

$$Q(E) = \sum_{i=1}^{m} \sum_{j=1}^{n} \left[\frac{eij}{sij} \right]^2 = \sum_{i=1}^{m} \sum_{j=1}^{n} \left[\frac{Xij - \sum_{K=1}^{P} gikfkj}{Sij} \right]^2 \quad (4)$$

where S_{ij} is the estimated uncertainty, n is the number of samples, and m is the number of species.

The dataset matrixes (200 samples × 25 PAHs) included concentrations and uncertainties of PAHs were loaded into the US-EPA PMF 5.0 model (US-EPA 2014). Then 100 bootstrap runs and a minimum correlation of 0.6 were performed to test the uncertainty and stability of the resolved profiles in the base run. A critical process in PMF modeling is the determination of the correct number of factors. The numbers of factors were chosen depending on the background understanding of the sources. During the PMF analysis, the model was run for different number of factors and finally, the six-factor solution gave the most stable results and the most easily interpretable factors. So, six factors (sources) of PAHs including diesel combustion, gasoline combustion, industrial activities, natural gas combustion, evaporative-uncombusted, and other sources were revealed by employing the model. The relevant source profiles of PAH species of the six factors related to the fall data have been shown in Fig. 2. The lowest or optimized Q value was selected as Q (robust) and Q (true) values were calculated including all points. The Q (robust) values for fall, winter, spring, and summer seasons were 945.240, 523.365, 355.038, and 309.242, respectively, and the correspondence Q (true) values were 952.040, 523.362, 354.968, and 309.193, respectively. More details

Fig. 2 Profiles of sources identified for PAHs in PM$_{2.5}$ (data of fall season) from the PMF model
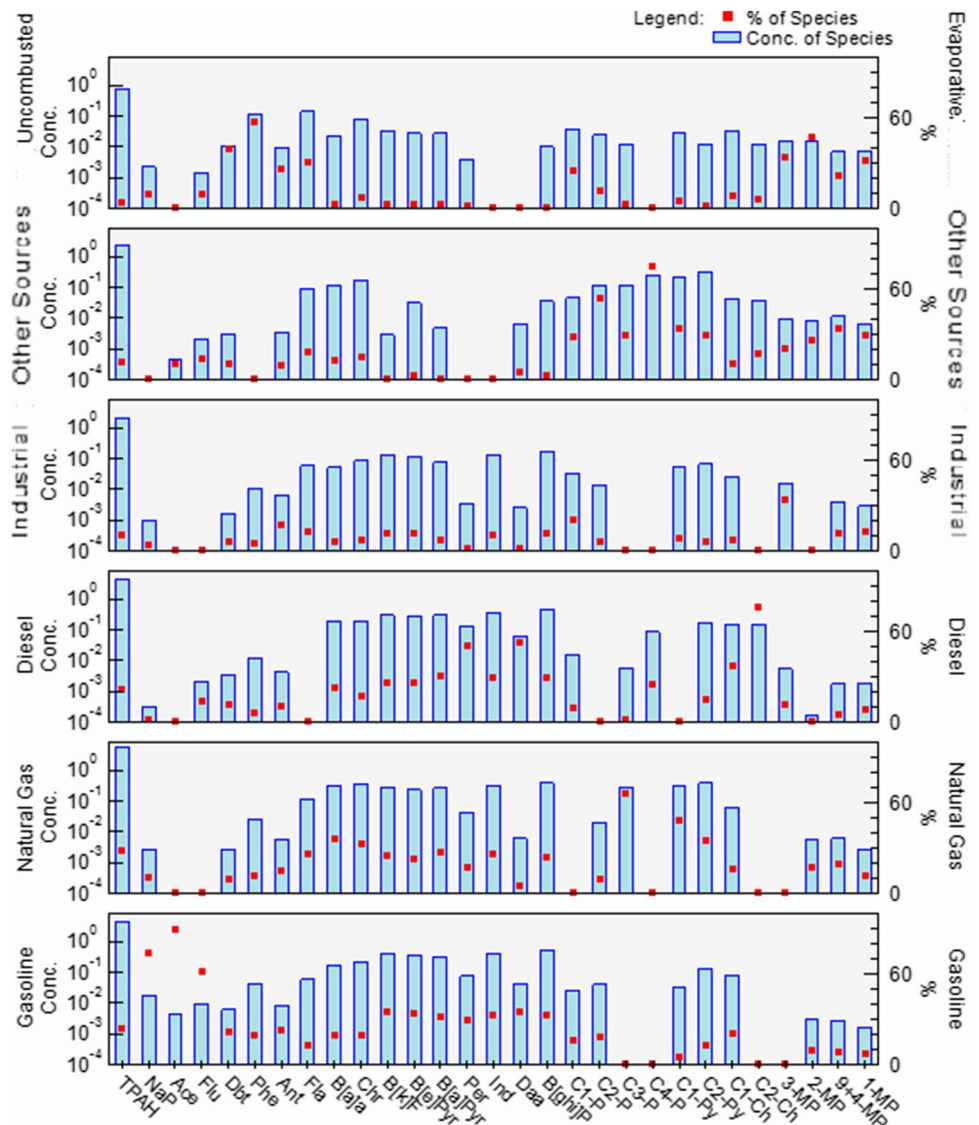
are provided in Table S5. Upon running the PMF analysis, rotation by changing Fpeak values between -1 and $+1$ was conducted, but no significant variations or improvements in source profiles were found.

Health risk assessment

Inhalation is one of the main human exposure pathways to PAHs. In this study, the toxic equivalency factors (TEFs) were used for quantitative risk estimate from inhalation pathway. TEF is an estimate of the relative toxicity of a chemical compared to a reference chemical. Benzo[a]pyrene as a reference compound is the most appropriate and widely used indicator to assess the carcinogenic potential of PAHs through different exposure pathways. Therefore, the Benzo[a]pyrene equivalent concentration (BaPeq) was estimated based on the following equation:

$$\sum BaPeq = \sum_{i}^{n=1} Ci \times TEFi \tag{5}$$

where C_i is the concentration of the ith target PAH compound (ng/m^3), TEF$_i$ is the toxic equivalency factor of the ith target compound. TEFi values of the PAHs were taken from the literature (Collins et al., 1998; Nisbet and Lagoy, 1992). The carcinogenic risk of each PAH as a lifetime lung cancer risk (LLCR) was calculated using Eq. (6).

$$LLCR = \sum BaPeq \times UR[BaP] \tag{6}$$

BaPeq was estimated by Eq. (5), where UR[BaP] was the inhalation cancer unit risk factor of BaP. The recommended value of UR [BaP] is 8.7×10^{-5} (WHO, 2000) showing the insidence of 8.7 cases per 100,000 people at risk of cancer from inhalation a BaP equivalent concentration of 1 ng/m^3 within their lifetime of 70 years.

Results and discussions

PM$_{2.5}$ and PAH concentrations

The concentrations of PM$_{2.5}$ and 19 individual PAH compounds in the collected samples were analyzed. Mean values of PM$_{2.5}$ concentrations and individual PAH compounds in the four seasons are shown in Table 1. PM$_{2.5}$ concentrations ranged from 8 to 291 μg/m^3 with a mean concentration of 60.21 ± 53.96 μg/m^3 and the total concentration of 19 PAH compounds ranged from 0.32 to 61.38 ng/m^3 with a mean concentration of 4.65 ± 8.54 ng/m^3.

The mean PM$_{2.5}$ mass concentrations in all seasons exceeded target value of 25 μg/m^3 proposed by the World Health Organization (WHO 2005), whereas the Iranian national standard value is 35 μg/m^3.

Table 1 Mean PAH concentrations (ng/m^3) in PM$_{2.5}$ samples collected in various seasons

Label	Fall	Winter	Spring	Summer	Annual mean	Range
Nap	0.029	0.012	0.002	0.001	0.011	N.D.–0.060
Acy	0.011	0.000	0.000	0.000	0.003	N.D.–0.044
Ace	0.005	0.002	0.000	0.000	0.002	N.D.–0.017
Flu	0.021	0.005	0.000	0.000	0.007	N.D.–0.074
Dbt	0.035	0.008	0.002	0.000	0.011	N.D.–0.176
Phe	0.240	0.043	0.000	0.000	0.071	N.D.–0.540
Ant	0.050	0.012	0.006	0.004	0.018	N.D.–0.113
Fl	0.610	0.106	0.033	0.031	0.195	0.010–2.322
Pyr	0.735	0.118	0.040	0.040	0.233	0.012–3.137
B[a]A	1.172	0.113	0.041	0.029	0.339	N.D.–5.540
Chr	1.306	0.176	0.060	0.040	0.395	0.008–4.848
B[b]F	1.139	0.224	0.130	0.103	0.399	N.D.–5.417
B[k]F	1.391	0.301	0.144	0.110	0.486	N.D.–6.220
B[e]P	1.262	0.306	0.173	0.123	0.466	0.027–5.164
B[a]P	1.357	0.228	0.099	0.048	0.433	N.D.–7.883
Per	0.383	0.059	0.017	0.002	0.115	N.D.–2.611
I[c]P	1.618	0.314	0.168	0.206	0.577	0.043–9.584
D[ah]A	0.203	0.031	0.015	0.009	0.064	N.D.–1.406
B[ghi]P	2.067	0.553	0.309	0.359	0.822	0.060–9.530
PAHs	13.631	2.609	1.240	1.106	4.647	0.318–61.380
PM$_{2.5}$ (μg/m^3)	140.675	26.790	30.102	43.270	60.209	7.987–291.157

N.D. non detected

Seasonal variations of the PAH concentrations during the sampling period showed the highest and the lowest concentrations in fall and summer, respectively. One of the main reasons of increasing concentration in fall season could be due to temperature inversion which usually enhance the diffusion of $PM_{2.5}$ and consequently affect PAH concentrations (Chen et al. 2020).

The dependence of PAHs concentration on atmospheric temperature and the increase in particulate PAHs concentration during the cold season have been reported by others (Chen et al. 2020; Vlachou et al. 2019; Tsapakis and Stephanou 2005; Li et al. 2006). Seasonal variations of emission sources and meteorological conditions such as temperature and inversion that influence the difference of gas-to-particle partitioning may result in a difference of PAHs concentration in cold and warm seasons (Tan et al. 2006). Reduced atmospheric dispersion as well as reduced photochemical reactions in low atmospheric temperatures can lead to higher pollutant concentrations during cold season (Chen et al. 2020; Vlachou et al. 2019), as it is seen for PAH concentrations in the cold seasons in comparison to the warm seasons (Table 1). In contrast, the increase in ambient temperature during the warm season may result in an increased evaporation of the particle phase of PAHs to the gas phase. This could be the reason of significant decrease of LMW PAH concentrations (e.g., Nap, Acy, Ace, Flu, Dbt, and Phe) more than 10 times in summer than the fall (Table 1). In addition, the PAHs degradation by photochemical or thermal reactions in the atmosphere especially in the warmer seasons is well known (Dörr et al. 1996). Furthermore, increasing the fossil fuel

consumption is another reason for the higher concentrations of PAHs in the cold season (Wu et al. 2014). It seems that increasing the fossil fuel combustion and temperature inversion during the cold seasons that influence the dispersion of air pollutants could have the main role of increasing the PAHs concentration. Since the particle-bound PAHs were analyzed in the current study and emission sources were mostly active throughout the year, meteorological conditions (e.g., inversion) might be the most important factor affecting $PM_{2.5}$ and PAH concentrations.

Table 2 shows the mean concentration of $PM_{2.5}$ and ΣPAHs in different cities worldwide. The mean concentration of $PM_{2.5}$ in Isfahan were greater than those in Tehran, Iran (Ali-Taleshi et al. 2020; Kermani et al. 2017); Zaragoza, Spain (Callén et al. 2014); Kanazawa, Japan (Xing et al. 2020); São Paulo, Brazil (Bourotte et al. 2005); Venice, Italy (Masiol et al. 2012); Islamabad, Pakistan (Mehmood et al. 2020); Thessaloniki, Greece (Tolis et al. 2015); and Taiwan, China (Chen et al. 2016), but lower than those in Anshan, China (Wang et al. 2020b); Beijing, China (Feng et al. 2018); and Guangzhou, China (Liu et al. 2015). The concentration of ΣPAHs in Isfahan was higher than those in Bangi, Malaysia (Khan et al. 2015); Zaragoza, Spain (Callén et al. 2014); Kanazawa, Japan (Xing et al. 2020); and Taiwan, China (Chen et al. 2016), but lower than those in Tehran, Iran (Ali-Taleshi et al. 2020; Taghvaee et al. 2018; Kermani et al. 2017); Jinan, China (Zhang et al. 2019); Seoul, Korea (Kang et al. 2020); Anshan, China (Wang et al. 2020b); Beijing, China (Feng et al. 2018); Thessaloniki, Greece (Tolis et al. 2015); Islamabad, Pakistan (Mehmood

Table 2 Annual mean concentrations of $PM_{2.5}$ and ΣPAHs in various locations

City	Location type	$PM_{2.5}$ concentration (μg/m^3)	PAHs concentration (ng/m^3)	No. of PAHs	References
Isfahan, Iran	Urban-Industrial	60.21	4.65	19	**This study**
Tehran, Iran	Urban	59.8	30.1	16	Ali-Taleshi et al., 2020
Tehran, Iran	Urban	41.19	7.92	16	Kermani et al., 2017
Tehran, Iran	Urban	No data	12.2	16	Taghvaee et al., 2018
Bangi, Malaysia	Semi-urban	No data	2.79	16	Khan et al., 2015
Anshan, China	Industrial/residential	81.22	131.98	16	Wang et al., 2020b
Beijing, China	Residential	136.9	58.30	20	Feng et al., 2018
Jinan, China	Urban	No data	39.8	19	Zhang et al., 2019
Zaragoza, Spain	Urban	13.05	2.14	19	Callén et al., 2014
São Paulo, Brazil	Urban	11.9	10.8	16	Bourotte et al., 2005
Venice, Italy	Industrial	32	10	11	Masiol et al., 2012
Seoul, Korea	Urban	No data	5.6	14	Kang et al., 2020
Kanazawa, Japan	Urban	16.1	1	9	Xing et al., 2020
Islamabad, Pakistan	Urban	51.59	25.69	16	Mehmood et al., 2020
Thessaloniki, Greece	Urban	37.75	7.02	16	Tolis et al., 2015
Taiwan, China	Urban	31.7	2.98	22	Chen et al., 2016
Guangzhou, China	Urban	64.88	33.89	17	Liu et al., 2015

et al. 2020); São Paulo, Brazil (Bourotte et al. 2005); Venice, Italy (Masiol et al. 2012); and Guangzhou, China (Liu et al. 2015).

Distribution of PAHs along seasons

According to the number of rings, PAHs are classified into five groups including 2-rings, 3-rings, 4-rings, 5-rings, and 6-rings. The analyzed PAHs in this study have been classified as follows: 2-rings (Nap); 3-rings (Acy, Ace, Flu, Dbt, Phe and Ant); 4-rings (Fl, Pyr, BaA and Chr); 5-rings (BbF, BkF, BeP, BaP, Per and DahA); and 6-rings (BghiP and Ind). They have been further divided into low molecular weight (LMW, 2- and 3-ring PAHs) and high molecular weight (HMW, 4-, 5-, and 6-ring PAHs). The concentration of high molecular weight (HMW) PAHs were significantly higher than of the low molecular weight (LMW) PAHs in all seasons (Fig. 3).

A similar composition pattern over the four seasons was observed with the highest contribution from the 5–6-ring (79.5%), followed by the 4-ring (18.5%) and the 3-ring PAHs (2%). The LMW PAHs can be formed in the pyrolysis of uncombusted fossil fuels, but the HMW PAHs mainly originate from high-temperature combustion processes such as vehicular exhaust (Dachs et al. 2002). Therefore, predomination of HMW PAHs in the study area indicated that PAHs mainly originate from combustion sources.

Since PAHs are semi-volatile organic compounds, they are partitioned between particle and gas phases. Low molecular weight PAHs tend to be more concentrated in the gas phase while the ones with higher molecular weight are often associated with particulates. So low concentrations of LMW PAHs in the samples of this study may just related to the particle-bound compounds not the total fractions including particle and gas phases.

Source apportionment of PAHs

Diagnostic ratios

The ratios of LMW/HMW, An/(An + Phe), (anthracene to anthracene plus phenanthrene), Flu/(Flu + Pyr), (fluoranthene to fluoranthene plus pyrene), BaA/(BaA + Chr), (Benzo[a]anthracene to Benzo[a]anthracene plus Chrysene), and IP/(IP + Bghi), (Indeno[1,2,3-cd]pyrene to Indeno[1,2,3-cd]pyrene plus Benzo[ghi]perylene) were used for identification of PAH sources. Figure 4 shows cross plots of PAHs ratios illustrating the different source types. Similar distribution patterns were observed along different seasons. For An/An + Ph, 0.10 is taken as a threshold to discriminate petrogenic from combustion sources. Values < 0.1 are usually considered as a petroleum source, while values > 0.1 indicate a combustion source (Yunker et al. 2002; Chen et al. 2013). The calculated ratio of An/(An + Ph) ranged from 0.09 to 0.34 with a mean value of 0.18. This means that the main source of PAHs in the city is from combustion process.

The Flu/Flu + Pyr ratio is another ratio that has been used to determine the emission source of PAHs. For this ratio, values less than 0.40 are distinctive of petroleum sources, values between 0.4 and 0.5 are distinctive of liquid fossil fuel combustion, and values above than 0.50 are distinctive of biomass or coal combustion (Yunker et al. 2002). These ratios have been also used to distinguish between gasoline and diesel emissions; where the values lower and higher than 0.5 refer to gasoline and diesel combustion, respectively. (Ravindra et al. 2006). In this study, the Flu/(Flu + Pyr) ratio was from 0.32 to 0.58 with a mean of 0.43. This shows the importance of gasoline and diesel emissions to Isfahan's atmosphere.

The ratio BaA/BaA + Chr is also declarative of the PAH sources. For this ratio, values lower than 0.20 indicate a petroleum source; the range from 0.2 to 0.35 suggests either
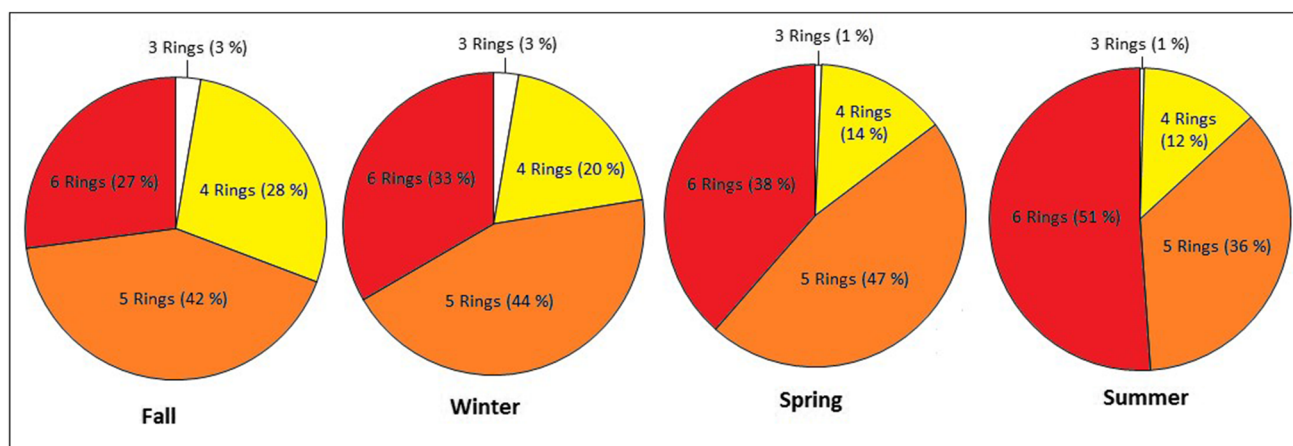
Fig. 3 Contribution of PAHs by number of rings (%) in the total PAHs in different seasons
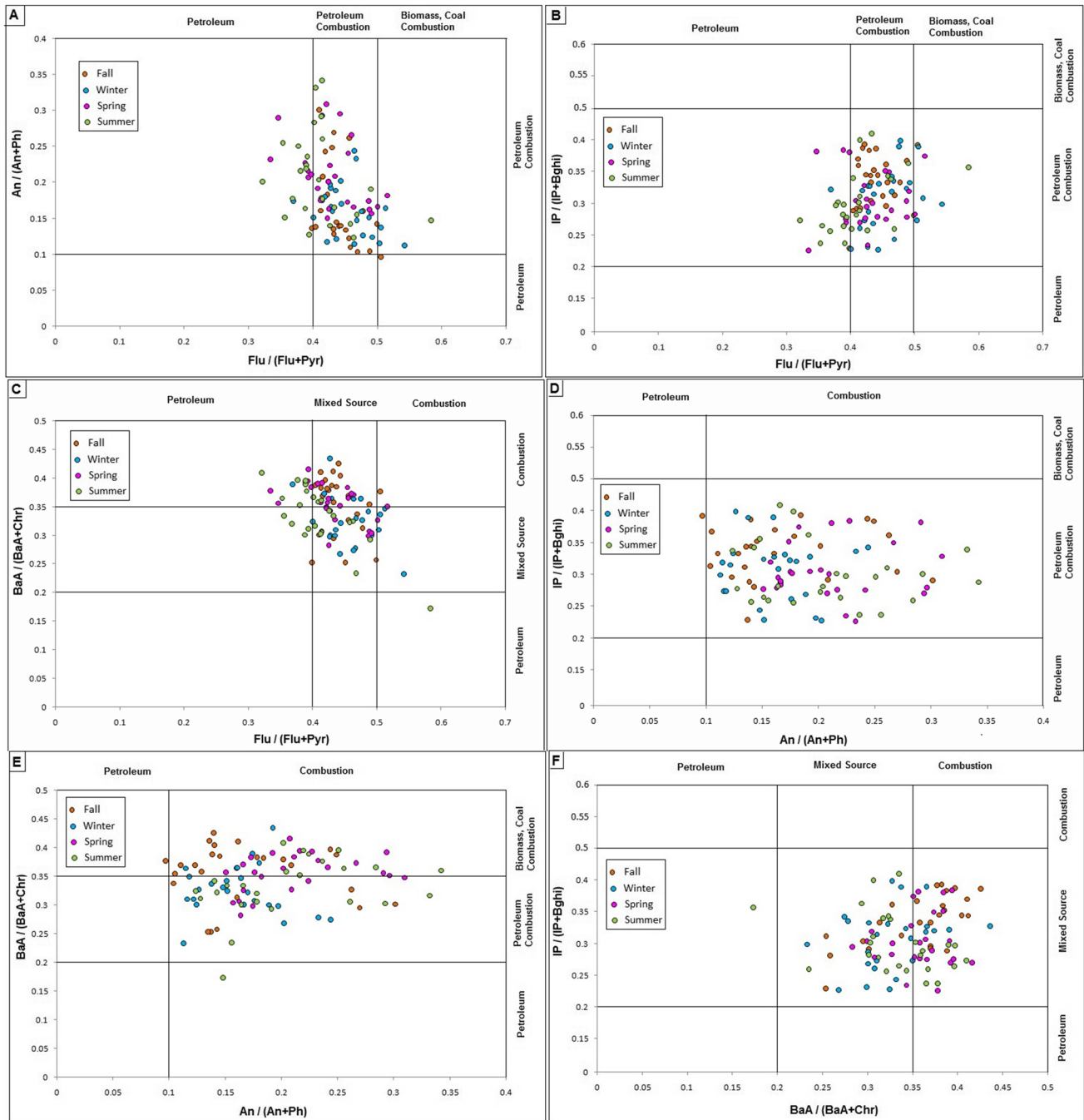
Fig. 4 Cross plots of various PAH ratios in PM$_{2.5}$ samples in Isfahan city, Iran

petroleum or combustion source and values higher than 0.35 imply a combustion source (Yunker et al. 2002; Akyuz and Cabuk 2010). Values for this ratio reported in the literature are 0.22–0.55 for gasoline, 0.38–0.64 for diesel (Simcik et al. 1997), and 0.5 for coal (Tang et al. 2005). The calculated ratio of BaA/(BaA + Chr) was from 0.17 to 0.43 with a mean of 0.34. This information reflects the significance of gasoline and diesel emissions to Isfahan city. Likewise, PAH sources may be identified by the ratio of IP/(IP + Bghi). For this ratio,

values lower than 0.20 indicate a petrogenic source; the range from 0.20 to 0.50 suggests liquid fossil fuel combustion, and values greater than 0.50 indicates grass, wood, or coal combustion (Yunker et al., 2002; Chen et al. 2005). For this ratio, some researchers documented 0.21–0.22 values for gasoline emissions (Rogge et al. 1993) and 0.35–0.70 for diesel emissions (Pio et al. 2001; Alves et al. 2001; Tang et al. 2005; Grimmer et al. 1983). In this work, the IP/(IP + Bghi) ratio

was from 0.22 to 0.41 with a mean of 0.31 which confirms the importance of gasoline and diesel emissions.

The ratio of LMW/HMW in all samples was less than 1 indicating overall combustion sources of PAHs. In conclusion of the PAH diagnostic ratios analysis, combustion of fossil fuels was found to be the main source of PAHs in ambient $PM_{2.5}$ in Isfahan.

Obviously, diagnostic ratios are a useful technique to identify the sources of PAHs. But, due to difficulty to distinguish between different emission sources, this technique should be used with a great caution. Furthermore, degradation (e.g., photolysis) and reaction with other species in the atmosphere can alter distribution pattern of PAHs during their transfer from emission sources to receptor sites (Ravindra et al. 2008; Pongpiachan 2014). There should be awareness that coals and fossil fuels from diverse origins and countries can generate different proportions of PAHs (Masclet et al. 1987). However, interpretation of PAHs sources must be done based on such numerical data and on the knowledge available about the historical background of the study area.

Source identification using PMF model

The predicted and measured PAH concentration showed a significant correlation ($r^2 = 0.99$, $p < 0.01$) (Fig. 5.). Regarding the factors used in PMF model, PAH compounds such as Flu, Chr, Pyr, BaA, BeP, BaP, Ind, and BghiP, which were related to industrial activities (Lin et al. 2011), were also considered in the current study to find the industrial sources. Among them, steel and iron industry can produce PAHs through

different processes such as sintering, casting and cooling, and coke manufacturing (Yang et al. 2002). Ciaparra et al. (2009) demonstrated that low and moderate molecular weight PAHs (Flu, Phe, Ant, FluA, and Pyr) explained by coke making and HMW PAHs (BaP, IcdP, DahP, DacP, and BghiP) arising from the sintering process. As several types of industries, including the steel and iron industries, power plants, brick, and cement factories are located around the city, therefore, we attributed this factor to "Industrial activities." LMW PAHs such as Nap, Flu, and Phe were introduced as markers of uncombusted petroleum (Liu et al. 2015, 2009; Marr et al. 1999). The higher contribution of this factor in the warm seasons and the association with LMW PAHs lead us to the conclusion that this factor arises from "evaporative-uncombusted" sources, mainly from the gasoline emissions from storage tanks and pumps in gasoline stations. HMW PAH compounds such as BghiP, Ind, Pyr, and BeP were reported as chemical tracers of gasoline combustion by some researchers (Khan et al. 2015; Guo et al. 2003; Schauer et al. 2002). Gasoline is widely used in Isfahan and gasoline-fueled cars and motorcycles have been found to be important sources of hydrocarbons in Isfahan. In recent decades, gasoline has been used primarily as a fuel in automobiles. Vehicles, especially those for private use, have increased since past few decades in Isfahan city. Due to ambiguous characteristics, the fourth factor was left as "other sources." A mixture of minor sources such as wood, biomass and coal combustion, and natural dust seems to explain this factor. The dominant PAHs in factor of "diesel combustion" were BkF, BaP, Ind, and BghiP which have been reported as
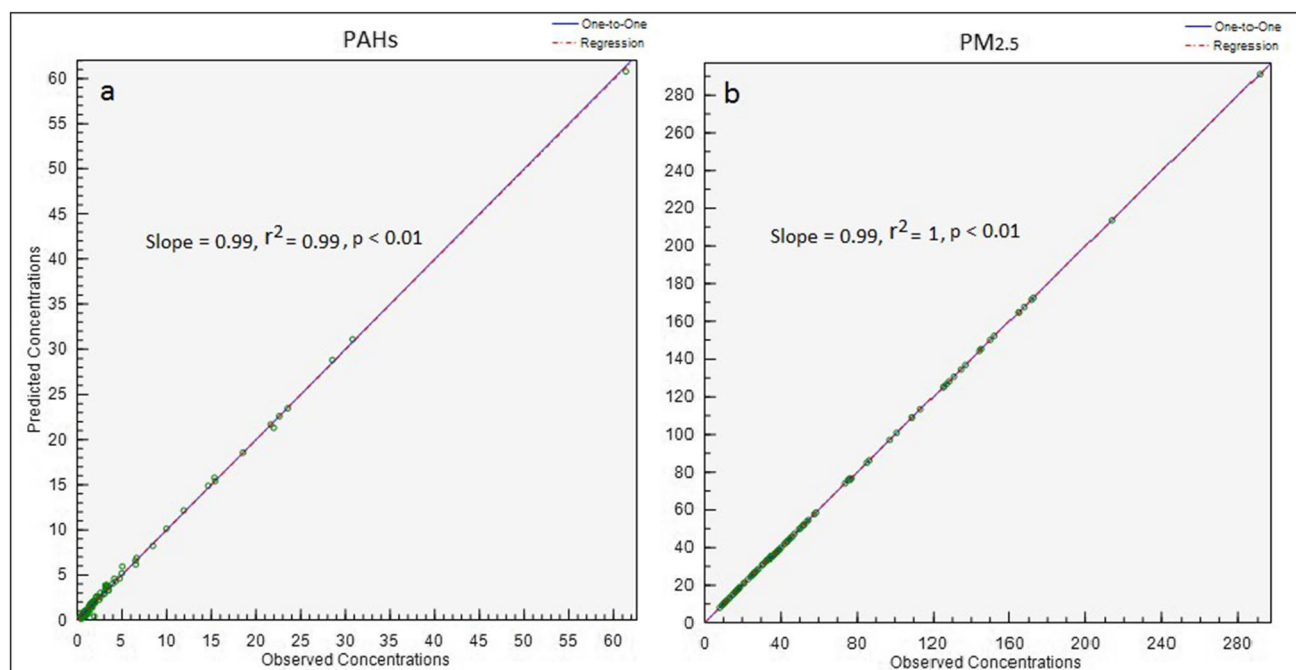
Fig. 5 Correlation between the PAH concentrations determined by PMF model and the observed values

the tracers of this source (Fang et al. 2016; Harrison et al. 1996; Wang et al. 2009). BbF, BkF, BaP, and Ind have been reported as chemical tracers of diesel combustion in numerous studies (Yang et al. 2013; Chen et al. 2011; Lin et al. 2011; Khalili et al. 1995; Ravindra et al. 2008; Ma et al. 2014; Harrison et al. 1996; Wang et al. 2014, 2015). Diesel fuel has been widely used in trucks and public transportation in Isfahan city. Moreover, the influence of power plants, and steel and iron industries using diesel as fuel may be also reflected in this factor. The natural gas is not only the main source of heating and cooking in Isfahan city, but also used as fuel in various industries and power plants through the region. So considering the markers such as Phe, Flu, BaA, Chr, and Pyr, reported by Jamhari et al. (2014), Khan et al. (2017), and Simcik et al. (1999), this factor named "natural gas combustion" was also used as one of the source factors of the model.

The contributions of each PMF factor to the level of total PAHs in different seasons are shown in Fig. 6. The source fingerprints in different seasons are shown in Fig. S1. For the cold seasons including fall (December–January months) and winter (February–March), the contributions were respectively included by natural gas combustion (28.4 and 9.5%), gasoline combustion (23.8 and 33.1%), diesel combustion (21.9 and 22.3%), industrial activities (10.3 and 9.9%), evaporative-uncombusted (3.9 and 9.5%), and other or unidentified sources (11.7 and 15.7%). The source contributions in spring and summer were gasoline combustion (29.3 and 26.0) diesel combustion (20.6 and 24.8%), evaporative-uncombusted (15.4 and 23.0%), natural gas combustion (14.1 and 15%), industrial activities (13.5 and 8.4%), and other or unidentified sources (7.1 and 2.8%), respectively. Since the natural gas are used in heating systems of residential-commercial sector and as a fuel in

various industries and power plants particularly in cold seasons (fall and winter), it showed the higher contribution in comparison to warm seasons (spring and summer). The natural gas is mainly used by industrial plants as fuel in spring and summer in the region. The total natural gas consumption of Isfahan city in 2018 was reported more than 3000 million m^3 where residential-industrial sectors consume more than 66 and 48% in cold (fall and winter) and warm (spring and summer) seasons, respectively (https://stat.mpo-es.ir). The power plants consume about 23 and 37% of the natural gas during cold and warm seasons, respectively (https://stat.mpo-es.ir). However, the consumption rate may change in different years. Since the main public transportation service in Isfahan city is provided by buses, they constitute together with trucks the major source of diesel combustion. It is estimated that more than 600,000 personal cars (as gasoline vehicles) and more than 1000 public transport buses (as diesel vehicles) are daily on road throughout the city (https://stat.mpo-es.ir).

To the best of our knowledge, no source apportionment studies have so far been performed to identify sources and quantify their contributions to ambient PMs in Isfahan. PAH source apportionment in respirable particles in Tehran, the capital of Iran, revealed five main sources including the following: diesel combustion (56.3%), gasoline combustion (15.5%), wood combustion and incineration (13.0%), industry (9.2%), and road soil particle (6.0%) (Moeinaddini et al. 2014). In other study in Tehran, source apportionment of $PM_{2.5}$-bound PAHs by PMF model revealed five main sources including the following: diesel exhaust (22.3%), unburned petroleum (15.6%), industrial (7.5%), gasoline exhaust (30.9%), and coal/biomass and natural gas combustion (23.6%) (Ali-Taleshi et al. 2020).
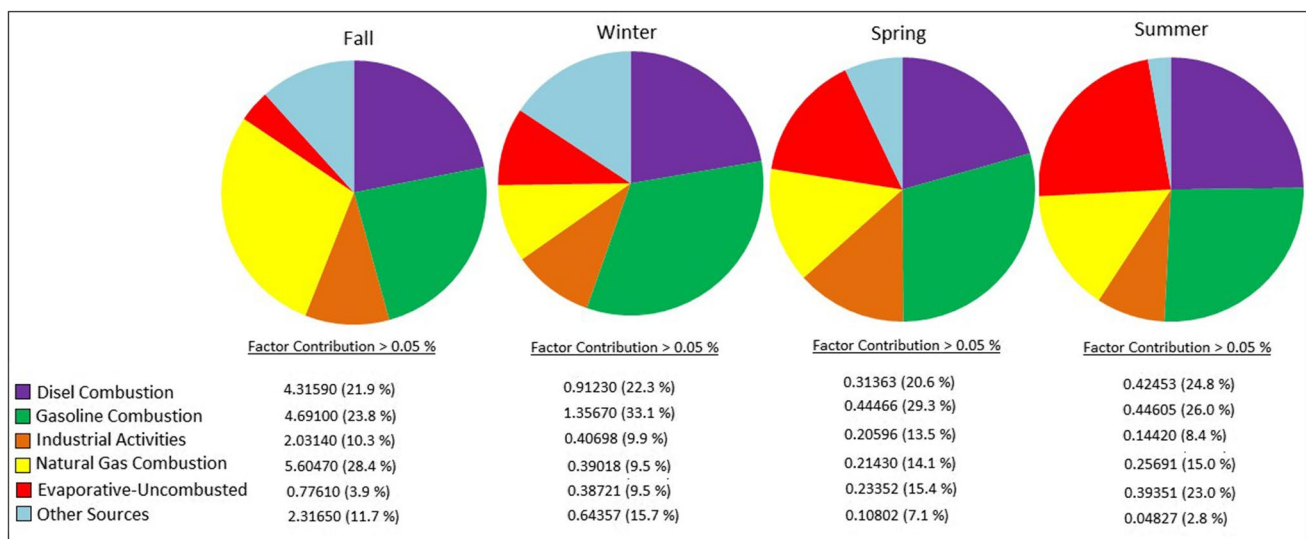

	Fall	Winter	Spring	Summer
	Factor Contribution > 0.05 %	Factor Contribution > 0.05 %	Factor Contribution > 0.05 %	Factor Contribution > 0.05 %
■ Disel Combustion	4.31590 (21.9 %)	0.91230 (22.3 %)	0.31363 (20.6 %)	0.42453 (24.8 %)
■ Gasoline Combustion	4.69100 (23.8 %)	1.35670 (33.1 %)	0.44466 (29.3 %)	0.44605 (26.0 %)
■ Industrial Activities	2.03140 (10.3 %)	0.40698 (9.9 %)	0.20596 (13.5 %)	0.14420 (8.4 %)
■ Natural Gas Combustion	5.60470 (28.4 %)	0.39018 (9.5 %)	0.21430 (14.1 %)	0.25691 (15.0 %)
■ Evaporative-Uncombusted	0.77610 (3.9 %)	0.38721 (9.5 %)	0.23352 (15.4 %)	0.39351 (23.0 %)
■ Other Sources	2.31650 (11.7 %)	0.64357 (15.7 %)	0.10802 (7.1 %)	0.04827 (2.8 %)

Fig. 6 Contribution of various sources in PAHs emission associated with $PM_{2.5}$ in different seasons by PMF model in Isfahan city, Iran

Health risk of PAHs

According to concentration of particle-bound B[a]P (Table 1), the concentration of this compound in fall was higher than the national standard value (i.e., 1 ng m^{-3}) of Iranian Department of Environment (Hoseini et al., 2016), while it was below the value in the other seasons showing the highest human exposure to PAHs and consequently the highest risk when inversion occurred (in December–January). The toxic equivalency factor (TEF), or relative potency, BaPeq of individual PAHs, and the lifetime lung cancer risk (LLCR) are shown in Table 3. The BaPeq of individual B[a]P accounted for more than 44% that shown by the total PAHs. Other studies reported that the carcinogenic potency of B[a]P was more than 50% (Khan et al., 2015), in the range of 81–85% (Cuadras et al., 2016) and in the range of 27–67% (Petry et al., 1996; Castellano et al., 2003) of the carcinogenic activity of total PAHs. The LLCR was in the range of 1.6×10^{-10} to 3.8×10^{-5} and for the overall data it was 8.5×10^{-5} which was very close to the critical value (8.7×10^{-5}), showing the cancer risk to the residents on exposure to PM$_{2.5}$-bound PAHs. However, the real PAH risk should be evaluated for both gas- and particle-phase PAHs in future researches and also should be investigated spatially and temporally for the people of different age groups living in the region.

Table 3 The BaP equivalents (BaPeq) and lifetime lung cancer risk (LLCR) of individual and total PAHs bounded to PM$_{2.5}$ samples in the study area

PAHs	Mean annual concentration (ng/m^3)	TEF[a]	BaPeq (ng/m^3)	LLCR
NaP	0.011	0.001	1.1×10^{-5}	9.6×10^{-10}
AceL	0.003	0.001	2.8×10^{-6}	2.4×10^{-10}
Ace	0.002	0.001	1.9×10^{-6}	1.6×10^{-10}
Flu	0.007	0.001	6.6×10^{-6}	5.7×10^{-10}
Phe	0.071	0.001	7.1×10^{-5}	6.2×10^{-9}
Ant	0.018	0.010	1.8×10^{-4}	1.6×10^{-8}
Fla	0.195	0.001	1.9×10^{-4}	1.6×10^{-8}
Pyr	0.233	0.001	2.3×10^{-4}	2.0×10^{-8}
B[a]a	0.339	0.100	33.9×10^{-3}	2.9×10^{-6}
Chr	0.395	0.010	3.9×10^{-3}	3.4×10^{-7}
B[b]F	0.399	0.100	39.9×10^{-3}	3.5×10^{-6}
B[k]F	0.486	0.100	48.6×10^{-3}	4.2×10^{-6}
B[a]P	0.433	1.000	432.8×10^{-3}	3.8×10^{-5}
Ind	0.577	0.010	5.8×10^{-3}	5.0×10^{-7}
Daa	0.064	0.100	6.4×10^{-3}	5.6×10^{-7}
B[ghi]P	0.411	1.000	422.1×10^{-3}	3.6×10^{-5}
Total PAHs	4.647	–	983×10^{-3}	8.5×10^{-5}

[a]Nisbet and LaGoy (1992)

Unit risk of BaP was 8.7×10^{-5} according to WHO (2000)

Conclusion

The annual mean concentrations of PM$_{2.5}$ and ΣPAHs were 60.2 ± 53.96 μg/m^3 and 4.65 ± 8.54 ng/m^3, respectively, in the study area. The mean PM$_{2.5}$ mass concentrations exceeded target value of the WHO standard. Source apportionment of PAHs using diagnostic ratios confirmed that the PAHs predominated by high molecular weight originate mainly from combustion sources especially fuel oil combustion. PAH source apportionment by PMF model revealed that gasoline and diesel combustion had about 50% contribution of PAHs in PM$_{2.5}$ showing the importance of air pollution control in transportation of the region. Furthermore, natural gas combustion mainly by residential-commercial and industrial sectors with contribution of about 9.5 to 28.4% in PAH production and also lifetime lung cancer risk of PM$_{2.5}$-bound PAHs revealed the importance of air pollution control in Isfahan Metropolitan. Although the source apportionment techniques used in this study are useful, however, both DRs and PMF model have uncertainties such as changing the distribution pattern of the compounds from the sources to the receptors and subjective interpretation of source profiles. Total PAHs in the atmosphere including gas and particle phase should be investigated in future studies, specially for the health risk assessments.

Author contribution Mohsen Soleimani: Conceptualization, methodology, resources, validation, writing original draft, writing-review and editing, supervision, project administration, funding acquisition.

Zohreh Ebrahimi: methodology, formal analysis, investigation, writing original draft, writing-review and editing.

Nourollah Mirghaffar: methodology, writing and editing, supervision,

Hossein Moradi: writing and editing, advisor.

Nasibeh Amini: methodology, formal analysis, investigation, validation, writing,

Kristoffer Gulmark Poulsen: methodology, validation, writing and editing.

Jan H. Christensen: resources, validation, writing-review and editing, funding acquisition.

Data availability The authors confirm that the data supporting the findings of this study are available within the article [and/or] its supplementary materials.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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