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PARTICULATE POLYCYCLIC AROMATIC HYDROCARBONS IN URBAN AIR OF TEHRAN

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ABSTRACT

Polycyclic Aromatic Hydrocarbons are ubiquitous in the urban atmosphere and one of the most atmospheric pollutants to be identified as suspected carcinogens .The objective of this study was to compare particulate PAH profiles from several sites across Tehran. Atmospheric concentrations of Polycyclic Aromatic Hydrocarbons in Tehran were evaluated to study particulate PAH profiles in different sites. The particles size distributions were unimodal with a fine mode diameter ($PM<0.49$ μm). Five sites were selected in residential areas and one site in rural area for representing various situations and role of motor vehicles in producing Polycyclic Aromatic Hydrocarbons. Organics on the filter were extracted with acetonitrile and a group of 16 Polycyclic Aromatic Hydrocarbons was selected for evaluation. PAH identification was done by High Performance Liquid Chromatography. Total PAH concentration associated with particles collected on the six-stage cascade impactor stages and after-filter in different sites ranged between 0.132 to 234.3 ng/m^3. The mean concentrations of Polycyclic Aromatic Hydrocarbons, collected on each stage were: 4.17 ng/m^3 for $PM_{10-7.2}$ μm, 5.69 ng/m^3 for $PM_{7.2-3}$ μm, 11.95 ng/m^3 for $PM_{3-1.5}$ μm, 9.18 ng/m^3 for $PM_{1.5-0.95}$ μm, 2.82 ng/m^3 for $PM_{0.95-0.49}$ μm and 47.91 ng/m^3 for $PM < 0.49$ μm respectively.

Key words: Polycyclic aromatic hydrocarbons, PM_{10}, size segregated, vehicle emissions

INTRODUCTION

Epidemiological studies have shown correlations between the concentration of particles in the air and increase of daily rates of respiratory and cardiovascular diseases and mortality (Douglas *et al.*,1992; Jonathan *et al.*,1995; Auron *et al.*,1995). Elevated lung cancer rates among women in rural china have been attributed to emissions from their coal-fired stoves (Mumford *et al.*,1987;Alder and Fischer,1994). Studies of populations occupationally exposed to diesel emissions data suggest that there is an association between this exposure and lung cancer (Roger,1987; Sharma and Patil, 1992a; Chow *et al.*, 2001). The carcinogenic activity of diesel emissions has also been demonstrated in rats (BCMELP, 1993; CEPA.,1994). In the late, Whitby and coworkers synthesized numerous measurements at atmospheric particle size distributions to describe the distribution of particle sizes in atmospheric aerosols (Seinfeld ,1986). Atmospheric particles are grouped into ultrafine, fine and coarse size modes. Sources of Polycyclic aromatic hydrocarbons (PAHs) in the urban atmosphere of industrialized countries include automobiles, re-suspended soils, refineries and power plants (Roger *et al.*, 1991; Venkataraman and Friedlinder, 1994). Tehran with population of more than 10 million has faced air pollution for a long time. Tehran occupies ~2300 km^2 at an elevation of 1200 m above mean sea level. It is bordered on the north, northwest, east and south-east by high to medium height (4000-1000m) mountain ranges. As in many large cities with limited ventilation, Tehran experiences air pollution problems, especially suspended particles. Particle size has important health implications, with the smaller particles having a greater ability to penetrate deeper into the airways and lungs of a person who is breathing in these particles (Kiss

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et al.,1998; Sanderson *et al.*, 2004). The composition and relative chemical load of different-sized particles is, therefore, clearly important in decreasing and determining risk. The human respiratory tract is an aerodynamic classifying system for airborne particles. The sampling device is used as a substitute for the respiratory tract as a dust collector or Impactor. The fraction of inhaled dust retained in the respiratory system and the site of deposition vary with size, shape, density and all the physical properties of the particles that constitute the aerodynamic dimensions (Roussel *et al.*, 1992). Polycyclic aromatic hydrocarbons are a group of organic compounds made up of two or more fused benzene rings in linear, angular or cluster arrangements (Counter,1990; Arey, 1998). Different arrangements of the rings have resulted in the identification of more than 200 different compounds. PAHs constitute a major group of carcinogens and mutagens in the environment (Chow *et al.*,1994a; Chen *et al.*, 2002). PAHs are readily inhaled into lung alveoli, posing a potentially significant health risk to humans (NRCC, 1993; CEPA ,1994). The objectives of this study were: 1) to characterize the concentration and particle size distribution of PAHs in ambient air ; 2) to quantify atmospheric PAHs at Tehran's atmosphere.

MATERIALS AND METHODS

Five sampling sites were selected to characterize the PAHs concentrations in ambient air in Tehran. On the whole, these sites were located in the North (Argentine square), South (Bahman square), East (Haft-houz square), West (Azadi square) and center of Tehran (Enghelab square). One more site was selected for evaluation and comparison of PAHs in the different sites. location of this site was in Tehran suburbs with low density traffic road way(Harm-Emam). Sixteen PAHs were measured simultaneously in particles phase of urban area in Tehran's atmosphere.

These compounds were Naphthalene (Nap), Acenaphtylene (Acy), Acenaphtene (Ace), Fluorene (Flu), Phenanthrene (Phen), Anthracene (Anth), Fluoranthene (Flt), Pyrene (Pyr), Benzo(α)Anthracene (BaA), Chrysene (Chr), Benzo[b]fluoranthene (Bbf),Benzo[k]fluoranthene (Bkf), Benzo[α]Pyrene (Bap), Dibenzo[ah]Anthracene (DBahA), Benzo[ghi]Perylene (Bghi) and Indeno(1,2,3-cd) Pyrene (Idp). In order to understand the distribution of PAHs at Tehran area, 1-year sampling program was undertaken. The size range of the particles collected was determined by the cascade impactors. Particles were collected using the six-stage Hi-Vol pressure impactor(HPI). The HPI was 50-percent efficiency cutoffs in aerodynamic diameters of 10, 7.2, 3, 1.5, 0.95 and 0.49 μm, for stages one through six, respectively. To obtain in role of motor vehicles in PAH emissions, sampling was done for 10-12 hours (8:00 am to 18:00 – 20:00 pm). Glass fiber filters were conditioned and weighed on a microbalance to determine the amount of particulate collected. After collection, filters were wrapped with aluminum foil and stored at 4°C in a refrigerator until they were sent to the laboratory for analysis.

Sample fractionation and PAH quantification

The samples analysis steps can be summarized as:

- Extracting analytes from collection media by sonication in acetonitrile for 30 min.
- Separating analyte fraction by High Performance Liquid Chromatography (HPLC).

The solvents used were acetonitrile and methanol 99.9 % purity (HPLC gradient Merck Company). Samples from the impactor stages and the after filter were split in half; then the half of filter was covered with approximately 2-5 mLof acetonitrile and extracted for 30 min in an ultrasonic bath (Unident model). Extracts were syringe-filtered through 0.45 μm millipore PTFE filters. The PAH species selected for analysis included in 2-6 rings (molecular weight of 128 – 278). The HPLC system was knaver equipped with an injector (20 μL) loop; the column (Restek) was 150 mm long, 3.2 mm inside diameter packed with 5 μm particle size of silica gel. A solvent gradient of acetonitrile and water was used with (40- 60 % acetonitrile) in 10 min, held at 100 % acetonitrile for 15 min at a flow rate 1.2 mL/min ; and a pressure of approximately 200 bar. Then the mobile phase composition was reset to initial condition. The HPLC system was calibrated using an external standard. A standard reference material (EPA 610

PAH mix-SUPELCO) was used at different dilutions to obtain calibration curves for each run. A good agreement between standard and sample chromatograms obtained on a given day . A fluorescence detector (RF-10AXL)was set at 280 and 390 nm wavelengths, respectively, for excitation and emission. The Fluorescence detector were unable to detect all of 16 species of PAH; therefore UV and Fluorescence detectors simultaneously used. The UV detector (K-2500) was operated at $\lambda = 254$ nm. The detectors were adjusted for maximum selectivity for each PAH. Prior to actual analysis, two or three injections of the liquid standard were made to ensure stabilization of the column.

RESULTS

The different PAHs were classified into two categories: low molecular weight (LM-PAHs, containing two to three ringed PAHs), and high molecular weight (HM-PAHs, containing four to six ringed PAHs) (Table 1).

Table 1: Mean of PAHs concentrations and their contribution to the total PAHs in PM10 from Tehran area

PAH Compounds	Molecular weight	No of Rings	Mean of Conc. (ng/m^3)	% of total PAHs
Low Molecular weight:				
Naphthalene	128.18	2	133.83	32.32
Acenaphtylene	152.2	3	157.56	38.05
Acenaphtene	154.2	3	36.93	8.92
Fluorene	166.23	3	20.99	5.07
Phenantherene	178.24	3	6.92	1.67
Anthracene	178.24	3	6.97	1.68
Total			363.21	87.72
High Molecular weight:				
Fluoranthene	202.26	4	15.97	3.86
Pyrene	202.26	4	2.07	0.50
Benzo[a]anthracene	228.3	4	5.34	1.29
Chrysene	228.3	4	5.00	1.20
Benzo[b]fluoranthene	252.32	5	4.78	1.15
Benzo[k]fluoranthene	252.32	5	2.05	0.50
Benzo[a]pyrene	252.32	5	5.53	1.34
Dibenzo[a,h]anthracene	278.35	5	1.80	0.43
Benzo[ghi]perylene	276.34	6	4.34	1.05
Indeno[1,2,3-cd]pyrene	276.34	6	3.95	0.95
Total			50.83	12.28

Particle mass concentrations and size distributions

Figs. 1-5 show the size distribution of PM10 at 5 sites vs. particles diameter(D_p). The particles size distributions were unimodal with a fine mode diameter PM<1 µm which has a long atmospherical residence time. Emissions from motor vehicles have been identified as a major contributor to ambient particulate matter.

PAH concentrations in the different particle size classes

Results of PAH size distributions measured at 5 sites were all unimodal are presented in Tables 2 to 6. In Table 7, comparison is made between PAHs concentration, in the 5 up mentioned sites and in the suburb of Tehran (Haram-Emam).

Of special concern are benzo[a]pyrene, benzo[b] flouranthene, benzo[J] flouranthene, benzo[k] flouranthene and Indeno[1,2,3-cd] Pyrene which have all been classified as probably carcinogenic to humans and defined as toxic compounds.

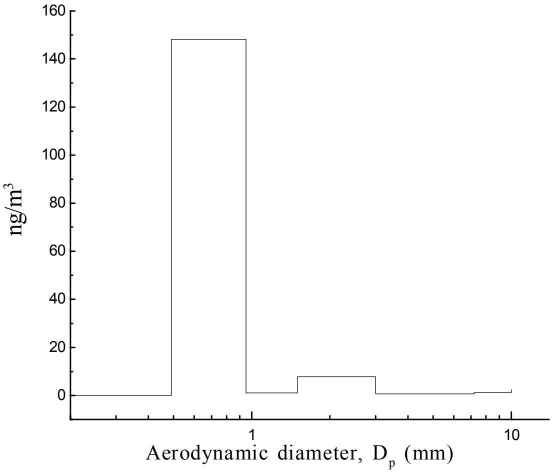

Fig. 1: Size distribution of PAH$_s$ at Enghelab Square

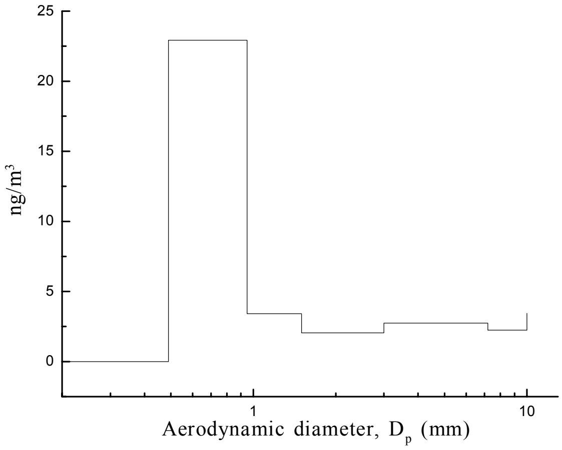

Fig. 2: Size distribution of PAH$_s$ at Argentine Square

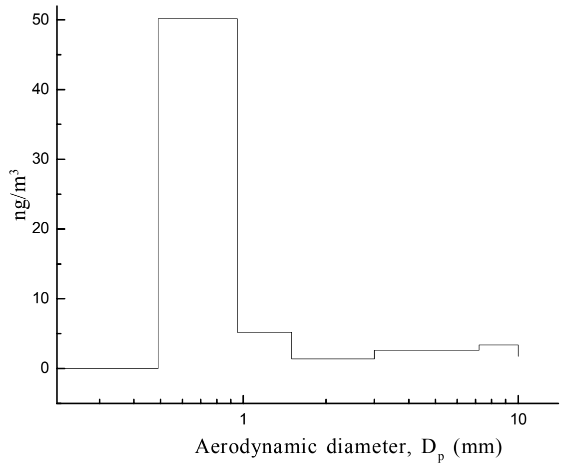

Fig. 4: Size distribution of PAH$_s$ at Bahman Square

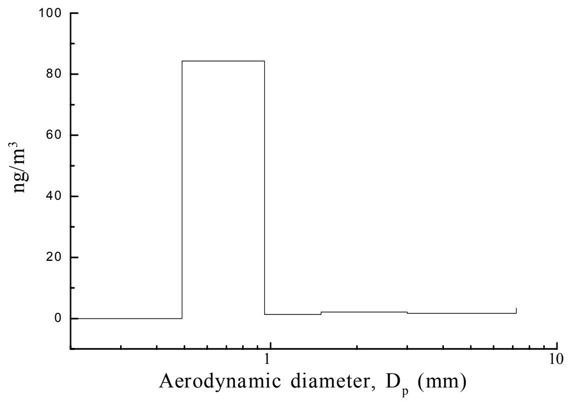

Fig. 3: Size distribution of PAH$_s$ at Azadi Square

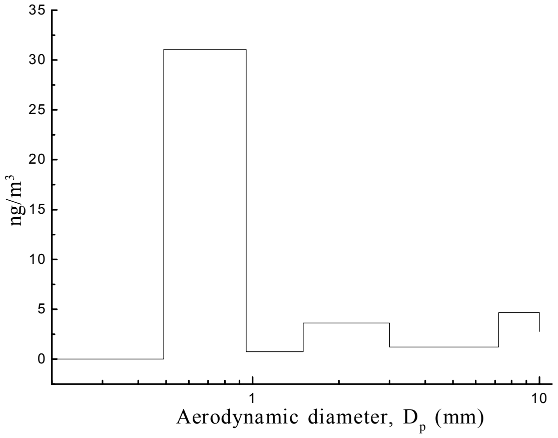

Fig. 5: Size distribution of PAH$_s$ at Haft-Houz Square

Table 2: The mean of PAHs concentration according to size distribution in Argentine square (ng/m^3)

PM size PAHs	10-7.2µm	3.0-7.2µm	1.5-3.0µm	0.95-1.5µm	0.49-0.95µm	<0.49µm
Naphthalene	3.24	---	---	---	---	---
Acenaphtylene	---	---	0.66	---	---	5.00
Acenaphtene	---	16.08	6.63	---	---	---
Fluorene	2.42	---	0.29	0.26	---	1.11
Phenantherene	0.17	1.56	0.74	0.23	0.51	7.02
Anthracene	---	1.26	0.05	0.05	---	---
Fluoranthene	0.22	0.13	0.06	0.78	---	---
Pyrene	0.02	0.08	0.05	0.04	0.05	0.42
Benzo[a]anthracene	0.24	0.07	0.15	0.36	0.91	1.29
Chrysene	0.17	0.33	0.41	0.53	0.28	1.92
Benzo[b]fluoranthene	0.05	0.07	0.09	0.17	0.18	1.27
Benzo[k]fluoranthene	0.03	0.15	0.04	0.05	0.09	0.74
Benzo[a]pyrene	0.03	0.05	0.04	0.05	0.11	1.13
Dibenzo[a,h]anthracene	0.03	0.07	0.14	0.03	0.11	0.26
Benzo[ghi]perylene	4.12	0.48	0.06	0.13	0.37	2.42
Indeno[1,2,3-cd]pyrene	---	0.32	0.07	0.06	0.12	1.24

PAH concentrations in the different particle size classes

Results of PAH size distributions measured at 5 sites were all unimodal are presented in Tables 2 to 6. In Table 7, comparison is made between PAHs concentration, in the 5 up mentioned sites and in the suburb of Tehran (Haram-Emam).

Of special concern are benzo[a]pyrene, benzo[b] flouranthene, benzo[J] flouranthene, benzo[k] flouranthene and Indeno[1,2,3-cd] Pyrene which have all been classified as probably carcinogenic to humans and defined as toxic compounds.

Table 3: The mean of PAHs concentration according to size distribution in Enghelab Square (ng/m^3)

PM size PAHs	10-7.2 µm	3.0-7.2 µm	1.5-3.0 µm	0.95-1.5 µm	0.49-0.95 µm	<0.49 µm
Naphthalene	---	---	---	2.53	---	---
Acenaphtylene	1.18	---	---	---	---	---
Acenaphtene	---	---	---	---	---	---
Fluorene	---	---	---	---	---	---
Phenantherene	0.52	0.21	1.07	2.10	9.46	46.68
Anthracene	0.23	1.17	1.98	---	2.76	---
Fluoranthene	---	---	---	---	---	---
Pyrene	0.05	0.21	0.23	0.08	0.14	5.82
Benzo[a]anthracene	0.13	---	0.40	0.06	0.06	1.96
Chrysene	---	0.43	---	---	0.03	1.28
Benzo[b]fluoranthene	0.11	---	0.26	---	---	---
Benzo[k]fluoranthene	0.03	0.23	0.03	---	0.04	---
Benzo[a]pyrene	0.06	0.10	0.11	0.19	0.09	2.63
Dibenzo[a,h]anthracene	0.08	0.16	0.19	0.01	0.07	1.62
Benzo[ghi]perylene	0.06	0.14	0.31	0.24	0.19	4.58
Indeno[1,2,3-cd]pyrene	---	0.19	---	0.09	0.17	3.83

Table 4: The mean PAHs concentration according to size distribution in Bahman Square (ng/m^3)

PM size PAHs	10-7.2 µm	3.0-7.2 µm	1.5-3.0 µm	0.95-1.5 µm	0.49-0.95 µm	<0.49 µm
Naphthalene	---	---	---	---	4.70	---
Acenaphtylene	---	---	---	---	---	---
Acenaphtene	---	---	---	---	---	---
Fluorene	---	---	---	---	---	---
Phenantherene	1.26	1.04	0.06	0.16	---	5.42
Anthracene	0.08	0.07	0.06	0.09	1.36	21.43
Fluoranthene	0.28	1.74	2.28	---	---	18.04
Pyrene	0.09	0.06	0.07	0.13	0.13	---
Benzo[a]anthracene	---	0.04	0.02	0.05	0.07	---
Chrysene	0.08	0.03	0.03	0.21	0.13	2.00
Benzo[b]fluoranthene	---	0.06	---	---	0.66	---
Benzo[k]fluoranthene	0.02	0.06	0.06	0.07	0.10	1.24
Benzo[a]pyrene	---	0.03	0.03	0.08	0.34	2.77
Dibenzo[a,h]anthracene	0.02	0.02	0.02	0.21	0.06	---
Benzo[ghi]perylene	0.05	0.06	0.06	0.50	0.39	6.80
Indeno[1,2,3-cd]pyrene	---	0.27	0.08	0.08	---	3.80

Table 5: The mean PAHs concentration according to size distribution in Haft Houz site (ng/m^3)

PM size PAHs	10-7.2 µm	3.0-7.2 µm	1.5-3.0 µm	0.95-1.5 µm	0.49-0.95 µm	<0.49 µm
Naphthalene	---	---	---	---	---	16.07
Acenaphtylene	---	---	---	---	---	10.76
Acenaphtene	---	---	---	---	---	---
Fluorene	1.48	---	---	---	---	---
Phenantherene	---	---	---	---	---	---
Anthracene	---	---	---	---	---	---
Fluoranthene	---	3.61	0.30	1.36	---	---
Pyrene	0.14	0.40	0.21	0.45	0.15	0.54
Benzo[a]anthracene	0.06	0.06	0.05	0.19	0.06	0.22
Chrysene	0.08	0.15	0.11	0.56	0.04	0.49
Benzo[b]fluoranthene	---	---	---	---	---	---
Benzo[k]fluoranthene	0.25	0.12	0.12	0.14	---	0.71
Benzo[a]pyrene	0.39	0.13	0.21	0.17	0.17	0.66
Dibenzo[a,h]anthracene	0.06	0.03	0.03	0.13	0.10	0.18
Benzo[ghi]perylene	0.31	0.15	0.17	0.36	0.21	1.45
Indeno[1,2,3-cd]pyrene	---	---	---	0.25	---	---

Table 6: The mean PAHs concentration according to size distribution in Azadi Square (ng/m^3)

PM size PAHs	10-7.2 µm	3.0-7.2 µm	1.5-3.0 µm	0.95-1.5 µm	0.49-0.95 µm	<0.49 µm
Naphthalene	---	1.03	---	---	---	---
Acenaphtylene	0.66	---	---	---	---	14.99
Acenaphtene	0.53	---	---	---	1.69	28.89
Fluorene	---	---	---	---	---	5.60
Phenantherene	0.79	0.64	0.75	1.13	0.65	2.17
Anthracene	---	0.08	0.23	0.13	0.33	20.13
Fluoranthene	---	1.27	---	---	---	---
Pyrene	0.07	0.10	0.08	0.09	0.11	4.38
Benzo[a]anthracene	0.03	0.05	0.06	0.06	0.15	4.42
Chrysene	0.07	0.07	0.08	0.06	0.05	2.43
Benzo[b]fluoranthene	0.26	0.10	0.29	0.24	0.34	4.03
Benzo[k]fluoranthene	---	---	---	0.01	0.05	1.66
Benzo[a]pyrene	0.06	0.15	0.10	0.09	0.19	3.24
Dibenzo[a,h]anthracene	0.06	0.05	0.07	0.08	0.11	1.18
Benzo[ghi]perylene	0.07	0.22	0.17	0.22	0.33	11.52
Indeno[1,2,3-cd]pyrene	---	---	0.90	0.14	0.45	9.59

Table 7: The mean PAHs concentration according to size distribution in Haram Emam station-suburbs area (ng/m^3)

PM size PAHs	10-7.2 µm	3.0-7.2 µm	1.5-3.0 µm	0.95-1.5 µm	0.49-0.95 µm	<0.49 µm
Naphthalene	---	---	---	---	---	---
Acenaphtylene	---	---	---	---	---	---
Acenaphtene	---	---	---	---	---	3.71
Fluorene	---	---	---	---	---	---
Phenantherene	---	---	---	---	---	0.63
Anthracene	---	---	---	---	---	---
Fluoranthene	---	---	---	---	---	---
Pyrene	0.04	0.05	0.15	0.04	0.04	2.50
Benzo[a]anthracene	0.01	0.01	0.03	0.02	0.05	0.64
Chrysene	0.03	0.01	0.03	0.04	0.03	0.65
Benzo[b]fluoranthene	---	---	---	---	---	---
Benzo[k]fluoranthene	0.04	0.05	0.04	0.06	---	---
Benzo[a]pyrene	---	---	---	---	0.05	1.83
Dibenzo[a,h]anthracene	0.03	0.02	0.02	---	0.02	0.55
Benzo[ghi]perylene	0.04	0.04	---	---	0.08	1.34
Indeno[1,2,3-cd]pyrene	---	---	---	---	---	---

DISCUSSION

Tehran urban areas with poor ventilation, unfavorable geographical and meteorological conditions, and a high number of emission sources including motor vehicles experience air pollution problems especially suspended particles. Air pollution in Tehran atmosphere are higher than the U.S. National Ambient Air Quality Standards (NAAQS) for particulate matter less than 10 µm (PM10). In all of stations mean of PM value < 10 µm are 5-10 times more than standards (annual mean 50 µg/m^3). As the tables detected, over 50 % of the mass were present in PM < 0.49 µm. Schonbuchner (2001) showed that ageing of aerosol during transport results in a shift of particle size from submicron towards larger particles of about 1 µm by growth processes. The second, smaller peak for 4-8 µm particles is probably caused by re-suspended soils mineral dust, plant tissue, pollen and refineries. The high aerosol concentration was associated with low average wind speed (1.5-2 m/s) and high frequency of calm conditions (60-80%), implying predominant anthropogenic sources, corroborated by the fine fraction of particulate (PM<0.5µm). Referring to Table 1, this study revealed that low molecular weight PAHs represented a main portion (87.6 %) of the total in airborne particles with 12.4 % high molecular weight PAHs. Results of PAH analysis indicated that existence of several of the higher molecular weight PAH$_s$ (Benzo(α)Anthracene, Chrysene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo[α]Pyrene), Benzo[ghi]Perylene, Indeno(1,2,3-cd) Pyrene confirm role of gasoline fueled emissions in Tehran's atmosphere. Energy use of residential buildings across Tehran (Iran) are dominated by natural gas with a small mount of electricity and heating oil. A major source of PAHs in Tehran is related to traffic and the number of gasoline and diesel vehicles. Emissions from motor vehicles have been identified as a major contributor to ambient particulate matter.

This study has shown that diesel exhaust is enriched in FLT,CHR and PYR, while gasoline exhaust has high BgP concentration.

Overaly, the atmospheric levels of ΣPAH for the sites in this study were Azadi (130 ng/m^3)> Enghelab (96.5 ng/m^3) > Bahman (79.1 ng/m^3) > Argentine (70.2 ng/m^3) > (44.1 ng/m^3)Haft Houz. Unfortunately at Azadi station in the west of Tehran area ,human carcinogenic compound like BaA (4.76 ng/m^3) , BbF (5.25 ng/m^3), BkF (1.72 ng/m^3), BaP (3.82 ng/m^3) and Ind (11.08 ng/m^3) were higher than other stations. Azadi station was largely influenced by traffic. One more site selected for evaluation and comparison of PAHs in the different sites. location of this site was suburbs with low density traffic road way(Harm-Emam).Concentration of PAH in this site found at 12.9 ng/m^3 (Table 7). The present study indicated that about 70% of PAH total content are on particles with diameter less than PM<0.49 µm (Azadi 87.9%; Bahman 77.7%; Enghelab 70.8%; Haft Houz 70.55; Argentine 34%). As mentioned before, Tables 2-6 show a typical PAH distribution for the study period, where the low molecular substances like Acy, Phen, Flu, Ace, Ant had a maximum load: (azadi-79.4%), (Enghelab-67.3%), (Arjantin-44%), (Bahman-31%) and (Haft Houz-12.23 %). Higher PAHs concentrations were proposed relative to irregular increased in consumption of fossil fuel combustibles; vehicle emissions and meteorological factors such as lower mixing layer height, low wind speed, scanty of rainfall and inversion events. Human carcinogenic compounds in this station were BaA (0.75 ng/m^3); BbF (0 ng/m^3); BkF (0.18 ng/m^3); BaP (3.82 ng/m^3); Ind (0 ng/m^3) and DahA (0.65 ng/m^3). It is interesting to add an evaluation of the health hazards to these conclusions, since the fraction PM < 2.5 µm is largely retained in the respiratory tract. With a normal respiratory rhythm, a man can be expected to sample 1200 m^3 air in about 3 months. Over this period in Tehran the human pulmonary system will be exposed to about 100.78 µg polyaromatic compounds (only through adsorbion on particles). During winter periods, the amounts of human carcinogenic PAHs will increase 3 fold up to 21.96 µg such as BaA (5.24 µg), BbF (4.92 µg). However health risk assessment of carcinogenic PAHs can not be related only to overall concentration because each PAH has a different carcinogenic potential. In Tehran, haphazard urbanization, unprecedented vehicular emissions and inadequate infrastructure development are supplementary factors for the decrease of air quality.

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