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Distribution Pattern and Contamination Levels of Some Polycyclic Aromatic Hydrocarbons Along Roadside Soil at Major Traffic Intercepts During Autumn in Jalandhar, India

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Autumn season study of some automobile generated polycyclic aromatic hydrocarbons (PAHs) along roadside soil was conducted at selected city locations in Jalandhar (Punjab), India to ascertain the contamination levels and their distribution behaviour in soil. The concentration and type of PAHs were determined at ten locations at 1, 2 and 3 m distances along roadside soil covering all major traffic intercepts within city. Samples were extracted in acetone and dichloromethane (1:1) using soxhlet extraction. The extracts were filtered on a micro silica gel column to remove impurities and the eluates obtained were injected to GC-FID. The city average concentration of PAHs was found to be 16.38 μg g^{-1} whereas, concentration of sixteen individual PAHs was found to vary between 0.01-252.55 μg g^{-1}. Average concentration of non-carcinogenic and carcinogenic PAHs was found 4.86 and 31.39 μg g^{-1} (ratio 1:6.45), respectively. Five ringed PAHs were found in highest concentration (around 40-70 %) whereas, two ringed PAHs were in minimum concentration (around 0.28-0.56 %) at most of the intercepts. Average highest concentration for any individual location was found 46.54 μg g^{-1} at location J4 at 1 m distance and minimum concentration was 0.77 μg g^{-1} at location J9 at 3 m distance from roadside. Benzo(a)pyrene was the individual PAHs observed in highest concentration at all intercepts vary between 1.77-252.55 μg g^{-1}. At most of the intercepts within city, total carcinogenic PAHs concentration was quite high (85 %) in comparison to non-carcinogenic PAHs (15 %). Comparison of the city average of PAHs with Indian metro cities and world data indicate that their concentration is equal or even higher.

Key Words: Polycyclic aromatic hydrocarbons, Roadside soil, Pollutants, Concentration, Jalandhar.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic chemicals consisting of two or more fused benzene rings. Polycyclic aromatic hydrocarbons are components of most fossil fuels and are ubiquitous in the natural environment[1]. Most environmental PAHs are products of incomplete combustion or pyrolysis of fossil fuels[2]. The lighter PAHs (2-3 rings), which are generally less carcinogenic, are mostly found in the gaseous phase while the heavier ones associate them with airborne particles. Heavier PAHs (> 3 rings) are rapidly attached to existing particles, usually soot particles by adsorption or condensation upon cooling of flue gases[3].

Roadside soil is one of the closed sink for automobile generated PAHs. They are deposited in soil after condensation and adsorption on particulate matter (soot or dust particle) in the air. It is a good indicator of pollution and environmental risk to human as it is continuously accumulated in soil from where it may come in human contacts in various ways depending upon seasons. Previous research showed that there is a significant level of PAHs found in soil all over the earth including remote areas which originated from forest fires or airborne pollution[4]. Soil from urban area usually consists of high concentration of PAHs, sometime 50 to 100 times higher than those in less populated and undeveloped areas. Urban areas are also reported to have higher soil concentration of PAHs than forest or agriculture soils, mainly because of direct exposure to vehicular emissions[5]. Once these PAHs enter in the soil, they accumulate in horizons rich in organic matter where they are likely to be retained for many years due to their persistence and hydrophobicity[6]. Soils are an important reservoir for these compound[7] and exchanges between soils and atmosphere is a widely studied process[8]. With the increase in fossil fuel combustion, resulting from the industrial expansion, traffic and population growth over last few decades, the atmospheric concentrations of PAHs in Asian countries are expected to be high[9].

Literature survey showed that a lack of soil PAHs studies in fast growing cities of India, where two, three and four wheelers with age varying between 1-20 years ply on the same road due to socio-economic condition. Lack of infrastructure development, maintenance, poor planning and socio economic condition has multiplied the problem further. Smaller vehicles (two three and four wheelers) run mainly on petrol and diesel fuels are major contributor of PAHs in city atmosphere (air and soil). Polycyclic aromatic hydrocarbons have drawn attention of the large number of scientific community due to their carcinogenic and mutagenic nature[10]. It is important to determine the type of PAHs in soil as their concentration in soil in various seasons very significantly and correlate with the corresponding levels in air[11].

In India most literature on PAHs in ambient air, roadside dust and soil is available mostly for metro cities. The objective of the present study is to determine the concentration, type, statical analysis of the results, distribution pattern and behaviour of PAHs (16 PAHs identified by EPA as potentional carcinogenic in the environment) during autumn in the road-side surface soil at 1, 2 and 3 meter distances from the road side soil at places of high/average traffic and population density areas within city environment of Jalandhar (a developing city) in the state of (Punjab) India.

EXPERIMENTAL

Punjab is one of the most prosperous and fast developing state of the India. City of Jalandhar is one of the major expending city of the state located in the centre of the Punjab situated between rivers Beas and Sutlej. The city, which has major road and rail connections, is a market for agricultural, textile, leather goods, wood products, hand tools and sporting goods. Jalandhar is situated at 710 31' east and 300 33' north at a distance of 146 kms from state capital Chandigarh on Delhi-Amritsar National Highway (Grand Trunk Road). According to 2001 Census provisional, the area of Jalandhar district is 3,401 km^2 and total population of the district was 19,53,508 persons. The city has shown tremendous growth in terms of population, infrastructure and traffic density during past decade. The climate of this district is on the whole years dry except during the brief south-west monsoon season (July to September). The average annual rainfall in the district is 703.0 mm.

Ten sampling locations (both public and commercial places within city) were chosen on the basis of high traffic density, urban populations and geographical dispersion. Major road intercepts chosen as sampling sites were Maqsuda (J1) , DAV (J2), Patel (J3), Workshop (J4) , Jyoti (J5), Mission (J6), BMC (J7), Bus Stand (J8), Nakodar (J9) and Football (J10) chowks.

Based on population and traffic density, above 10 locations were divided into; places of high population and traffic density *i.e.*, site 1 (where traffic was high but slow during 9 am to 9 pm) and places of average traffic and population density *i.e.*, site 2 (places with normal traffic speed and average population density during 9 am to 9 pm). Fig. 1 shows the road map of sampling sites and their location codes.

Sampling period: The soil samples were collected during autumn in the mid of October, 2009. A total of 30 samples were collected from ten major locations (J1-J10). The maximum, minimum temperature during the time of sampling was 25-5 ºC whereas; humidity was 20 % (minimum) to 80 % (maximum).

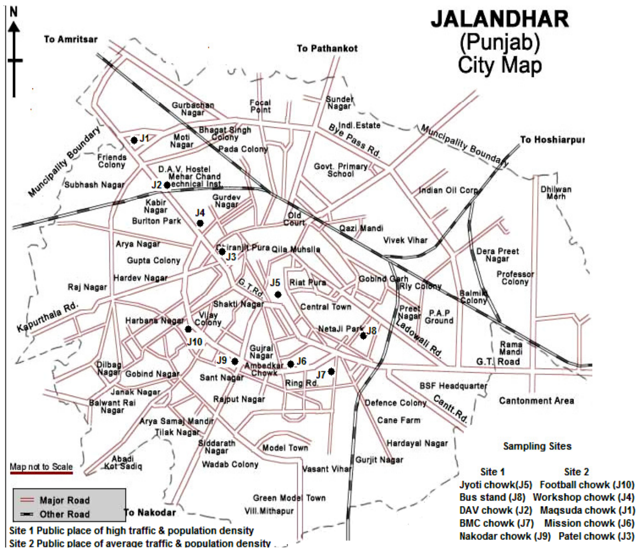


Fig. 1. City map of Jalandhar showing sampling locations

Sampling technique: From each sampling location three soil samples were collected from 1, 2 and 3 meter distance from different directions of the road (left and right side). Samples were collected from 6 cm below the surface layer. The bulk samples collected from sites were stored in polyethylene bags and brought to laboratory. Stones, leaves, debris and other extraneous materials were sieved out before analysis. All the samples were dried in a hot air oven maintained at 40-60 ºC for 24 h for removal of free moisture. The samples were preserved in desiccators and stored in dark before extraction.

Analytical grade reagents, solvents and standards used in the study were procured from Merck specialty Chemicals, Pvt. Ltd. Mumbai, India. PAH standards used in GC studies were procured from Dr. Ehrenstorfer GmbH Chemicals, Merck, Germany.

All glasswares of Borosil make glass (A grade) were used in the study. Glasswares were dipped in dilute nitric acid and washed with plain and double distilled water every time before use.

Polycyclic aromatic hydrocarbons present in the samples were analyzed on Nucon make microprocessor based gas chromatograph, (Model No 5765) using RH-5 capillary column (30 m length, 0.53 ID × 3.0 µm). Rotavapor and soxhlet extraction assemblies were used for extraction of PAHs from the soil samples.

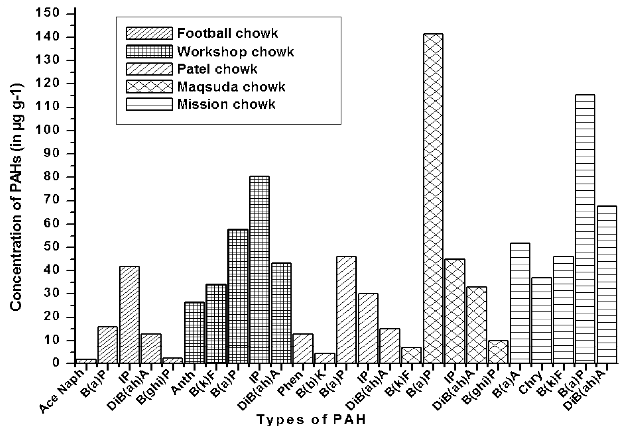
Extraction of polycyclic aromatic hydrocarbons: About 20 g of pre dried soil samples were heated at 40-60 ºC to remove any trace of moisture before extraction. Dried soil samples were transferred in soxhlet apparatus and extracted using acetone and dichloromethane (1:1 ratio) as solvents at the rate of 3 cycles/h for 8 h. The extract was allowed to cool and filtered through a Whatman filter paper No. 41. The filtrate was concentrated to 1 mL volume using rotary evaporation at 60 ºC under gentle vacuum. Extract were finally filtered through micro silica gel column of 6 cm length to remove impurities. The extracts collected were preserved in amber coloured sample tubes and stored in refrigerator below 4 ºC till analysis was carried out. Samples were later analyzed for types and concentration of PAHs by GC.

RESULTS AND DISCUSSION

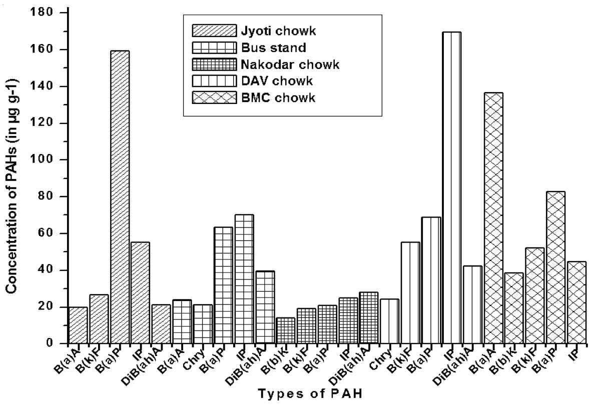
The results of average concentration of PAHs determined in 30 soil samples during autumn at different distances from roadside soil has been shown in Table-1. Ten sampling location were divided into high/average population and traffic density areas, categorized as site 1/site 2 has been shown in Table-1 and Fig. 1. In present study, 16 EPA identified PAHs namely napthalene (nap), acenapthene (ace nap), acenapthylene (ace naph), fluorene (flu), phenanthrene (phen), anthracene (anth), fluoranthene (flan), pyrene (pyr), benzo(a)anthracene (B(a)A), chrysene (chry), benzo(b)fluoranthene (B(b)F), benzo(k)-fluoranthene (B(k)F), benzo(a)pyrene (B(a)P), indeno(123cd)-pyrene (IP), dibenzo(ah)anthracene (Dib(ah)A) and benzo(ghi)-pyrelene (B(ghi)P) were determined.

Behaviour of PAHs at places of high population and traffic density intercepts: Concentration and types of PAHs at various intercepts were found to differ from place to place. Graphical presentation of concentration and types of five major PAHs detected at various locations (average within 3 m distance) are shown in Fig. 2a. It was observed from the figure that indeno(123cd)pyrene and benzo(a)pyrene were the PAH present in highest concentration varying between 25.07-169.62 and 20.98-159.51 μg g^{-1} in almost all intercepts, respectively. Benzo(a)pyrene was an individual PAH found in highest concentration (252.55 μg g^{-1} at 1 m) at location J1.

Average concentration of 16 PAHs at 1-3 m ($\bar{x}$ 1 m, $\bar{x}$ 2 m, $\bar{x}$ 3 m) was determined and the results obtained are shown in



(b)



(a)

Fig. 2. Distribution of five major PAHs and their concentration in soil (within 3 m) at (a) high traffic and population density and (b) average traffic and population density intercepts

Table-1. Graphical presentation of the results of variation of average concentration with distance is shown in Fig. 3a. The results shown in Fig. 3a indicate that the average concentration of 16 PAHs decreases with distance from roadside in most cases from 1-3 m. The observed behaviour was perhaps due to close proximity of intercepts from roadside in the form of buildings, plants, shops at high population and traffic density areas. Table-1 show that the average highest concentration of 16 PAHs among five locations between three distances was 46.46 μg g^{-1} at 1 m from roadside at location J7. The observed value was highest in roadside soil in any other fast developing or metro city in India.

Fig. 4a indicates average concentration of 16 individual PAHs within 1-3 m distance at five different intercepts. It was observed from the graph that indeno(123cd)pyrene and benzo(a)pyrene were two individual PAHs found in highest concentration at most intercepts. The lowest and highest concentration of these PAH was found to vary between 25.07-169.62 and 20.98-159.51 μg g^{-1}, respectively at these locations. Location J2 was an intercept surrounding by five-six educational institutes, where, thousands of student were exposed to high percentage of PAHs. The result also indicates that the concentration of PAHs varies with season and meteorological conditions.

TABLE-1
AVERAGE CONCENTRATION OF PAHS (16 PAHS) IN SOIL AT 1-3 m DISTANCE FROM ROADSIDE AT TRAFFIC INTERCEPTS OF HIGH (SITE 1) AND AVERAGE (SITE 2) TRAFFIC AND POPULATION DENSITY AREAS

S. No.	Sampling locations		Average Concentration of 16 PAHs in μg g^{-1} at different distances from roadside (m)			Average
			1 M	2 M	3 M	
1	Jyoti Chowk (J5)	Site 1	39.96	7.89	13.81	20.55
2	Bus Stand (J8)		25.26	16.90	13.27	18.51
3	DAV (J2)		41.5	21.08	19.93	27.50
4	BMC Chowk (J7)		46.46	11.74	11	23.06
5	Nakodar Chowk (J9)		15.80	10.34	0.77	8.97
6	Football Chowk (J10)	Site 2	3.03	11.06	2.32	5.47
7	Workshop Chowk (J4)		46.54	11.65	3.29	20.49
8	Patel Chowk (J3)		6.70	4.08	11.18	7.32
9	Maqsuda Chowk (J1)		13.05	11.68	21.87	15.53
10	Mission Chowk (J6)		45.25	26.30	10.99	27.51
11	Average		94.51	44.24	36.14	31.62

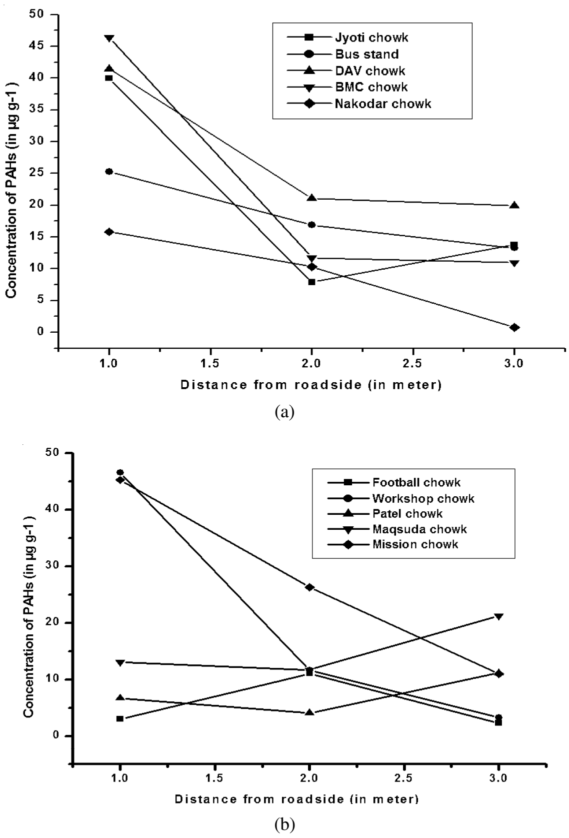


Fig. 3. Variation of average PAHs concentration (16 PAHs) in roadside soil as a function of distance in major traffic intercepts at (a) high traffic and population density and (b) average traffic and population density area

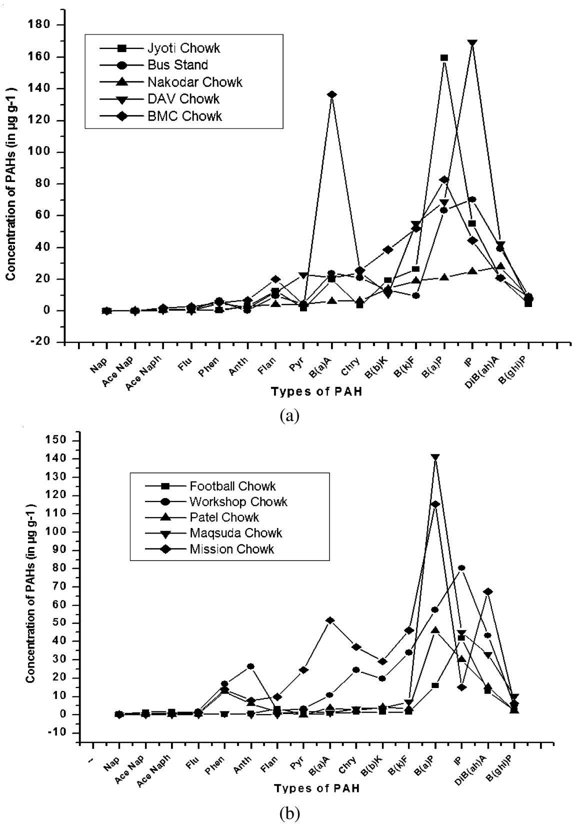


Fig. 4. Average concentration of 16 PAHs (within distance of 3 m) at major traffic intercepts in roadside soil at (a) high traffic and population density areas and (b) average traffic and population density areas

Ringwise distribution pattern and ratio of carcinogenic and non-carcinogenic PAHs at high traffic and population density intercepts: Results of ringwise distribution pattern of 2, 3, 4, 5 and 6 ringed PAHs (average concentration within 3 m) is graphically shown in Fig. 5a. The ring pattern of 16 PAHs in the Fig. 5a indicate that five ringed PAHs (B(a)P, B(k)f, B(b)f and DiB(ah)A) were present in highest concentration at most intercepts (40-70 %) whereas, two ringed PAHs were in lowest concentration (0.50 %) at all most all intercepts. In general the ringwise order of their concentration (%) was 5 > 6 > 4 > 3 > 2. EPA has classified seven PAHs as probable human carcinogens *i.e.*, B(a)P, B(k)f, B(b)f, DiB(ah)A, B(a)A, IP, B(ghi)P)[12,13]. Among seven PAHs identified by USEPA as potential carcinogens; all the seven PAHs were observed in the current area of the study. Out of seven, four PAHs are five ringed, two are five ringed and one of them was four ringed PAH. Ratio of carcinogenic and non-carcinogenic PAHs (percentage in total) and their average concentration (within 1-3 m) observed in the study is shown in Fig. 6a. It was observed from the figure that total percentage of probable human carcinogenic PAHs at all intercepts were quite high (85 %) in comparison to non-carcinogenic PAHs (15 %). The ratio of carcinogenic and non-carcinogenic PAHs was found to be quite high *i.e.*, approximate 6:1 at most locations.

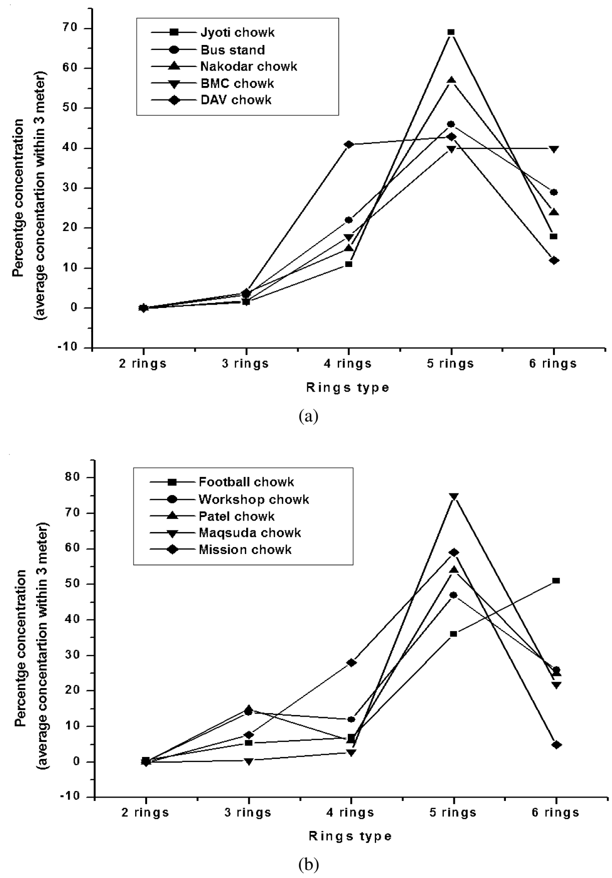


Fig. 5. Ringwise distribution pattern of 16 PAHs (average concentration within 3 m) identified at (a) high traffic and population density and (b) average traffic and population density intercepts

The high ratio of carcinogenic and non-carcinogenic PAHs at most intercepts in any fast developing city was the sign of potential risk to the residence of the city.

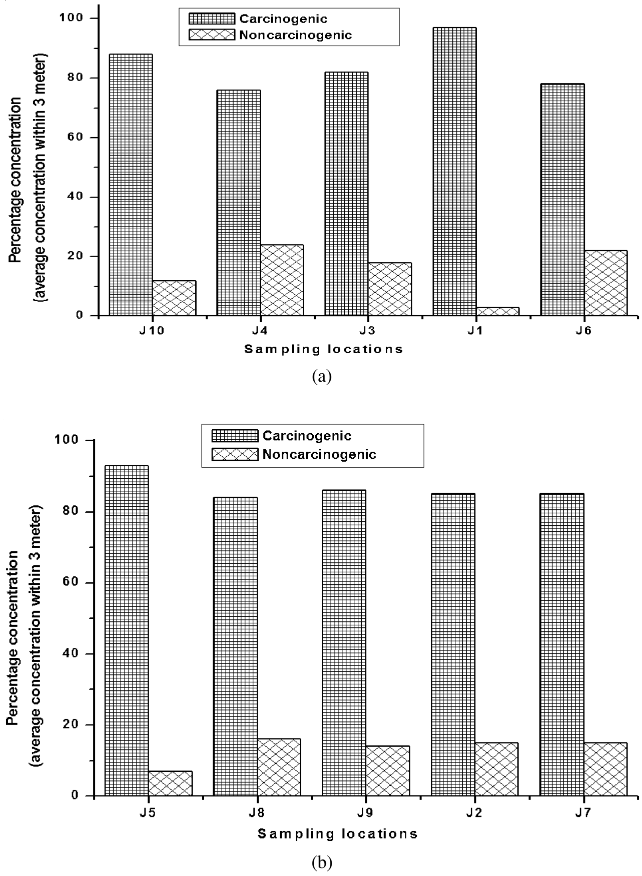


Fig. 6. Distribution behaviour (percentage in total) of carcinogenic (seven) and non-carcinogenic (nine) PAHs (a) average (within 3 m) at high traffic and population density intercepts and (b) average (within 3 m) at average traffic and population density intercepts

Behaviour of PAHs at places of average population and traffic density intercepts: It was observed that the concentration of PAHs and their types at various intercepts of average population and traffic density intercepts also vary from place to place. Graphical presentations of concentration of five major PAHs detected at various intercepts (average within 3 m) are shown in Fig. 2b. It was observed from the figure that DiB(ah)A and B(a)P were present in highest concentration varying between 12.73-67.59 and 16.02-141.47 μg g^{-1}, respectively (which was lower than site-1) results at most intercepts. B(a)P was an individual PAH found in highest concentration (252.55 μg g^{-1}) at 3 m distance from roadside at location J1. B(a)P was common PAH observed at both intercepts at highest concentration, whereas, DiB(ah)A was different from site 1 locations.

Results of average concentration of 16 PAHs within 1-3 m distance ($\bar{x}$ 1 m, $\bar{x}$ 2 m, $\bar{x}$ 3 m) is shown in Table-1 whereas, graphical presentation of the results of concentration variation with distance is shown in Fig. 3b. Average highest concentration of 16 PAHs among five locations (within 3 m) was observed 46.54 μg g^{-1} at 1 m at location J4. The results shown in Fig. 3b indicate that the average concentration of 16 PAHs decreases from 1-2 m and there after increase in most cases. Larger open area along roadside at these intercepts was responsible for different dispersion, suspension and distribution behaviour of PAHs at these intercepts. Average concentration of any individual PAHs within 1-3 m and their concentration are graphically shown in Fig. 4b. It was observed from this graphs that DiB(ah)A and B(a)P were two individual PAHs present in highest concentration in almost all intercepts. The lowest and highest concentrations of these PAHs were found varying between 12.73-67.59 and 16.02-141.47 μg g^{-1}, respectively. The value observed was comparatively lower than that of high traffic and population density intercepts. Temperature of soil was an important factor in determining the mobility of soil PAHs. Autumn seasons of sampling region receives high temperature ranging from 35-48 ºC. During autumn city environment remains dry and due to high traffic density the environment experience high concentration of particulate matter which adsorbs many volatile and non-volatile PAHs and settles in nearby area along roadside, results higher concentration during autumn.

Ringwise distribution pattern and ratio of carcinogenic and non-carcinogenic PAHs at average traffic and population density intercepts: Results of ringwise distribution pattern of 2, 3, 4, 5 and 6 membered ring (average percentage concentration in total within 3 m) is graphically shown in Fig. 5b. It was observed that five ringed PAHs (B(a)P,B(k)f, B(b)f and DiB(ah)A) were in highest concentration (40-70 %) whereas, two ringed PAHs were in lowest (0.28-0.50 %) concentration in almost all intercepts. In general ringwise increasing order of concentration (percentage in total) of various PAHs at these intercept was 5 > 6 > 3 and 4 > 2. Ratio of carcinogenic and non-carcinogenic PAHs (percentage in total) and there average concentration observed (within 3 m) in the study is shown in Fig. 6b. Fig. 6b shows that total percentage of probable human carcinogenic PAHs at these intercepts was quite high (85 %) as compared to non-carcinogenic PAHs (15 %). The ratio of two type of PAHs was found to be quite high *i.e.*, 6:1 at all locations.

Statistical analysis of results: In order to find extent out of human exposure to PAHs from roadside soil at high and average population and traffic density intercepts of the city the ratio of carcinogenic and non-carcinogenic PAHs was determined at various intercepts. Average concentration of non-carcinogenic and carcinogenic PAHs (within 3 m) at the place of high traffic and population density intercepts was 4.98 and 40.43 μg g^{-1} and their ratio was approximately 1:8 whereas, at the points of average traffic and population density intercepts the average value of non-carcinogenic and carcinogenic PAHs was 4.74-22.35 μg g^{-1} and their ratio was nearly 1:5. City average (for all ten intercepts within 3 m) of non-carcinogenic and carcinogenic PAHs was found 4.86 and 31.39 μg g^{-1}. The city average concentration of carcinogenic PAHs was 6.5 times higher in most places than non-carcinogenic PAHs.

Ratio of high and low carcinogenic PAHs was higher in autumn. Although there was no toxicity limit fixed for PAHs along roadside soil in India, the high and low carcinogenic PAHs results seems to be significant due to high ratio of these PAHs in roadside soil. Due to no PAHs limiting values in soil in India, the present data was compared with Mexican standards (0-6 μg g^{-1}), Polish standards (0.02-10 μg g^{-1}) and world wide data given in Table-2.

In order to compare the city average concentration of 16 PAHs of Jalandhar city with other cities of India, two tailed t-test was applied to the data. The results of the test applied to Jalandhar city average (16.38 μg g^{-1} current study), a developing

TABLE-2
WORLDWIDE SOIL PAH CONCENTRATION (ROADSIDE URBAN COMPILED FROM LITERATURE DATA)

Sr. No	Study area	PAH concentration (μg g-1)	Reference
1	Australia	3.30	Yang *et al.*[10]
2	USA	58.68	Rogge *et al.*[16]
3	Kota Bharu Malaysia	1.45	Fadzil *et al.*[15]
4	Mexican standards and polish standards	0-6, 0.02-10	Skrbic *et al.*[11]
5	Agra, India	12.9	Masih *et al.*[9]
6	Delhi airport soil data, India	4.43	Sharmila *et al.*[14]
7	Jalandhar, India	16.53	Present study

city to that of New Delhi airport soil data (4.43 μg g^{-1}) a metro city[14], indicate that the data of two places differ significantly at 5 % level of significance. This shows that PAH contamination level and distribution pattern (hence exposure to human being) of two places was quite different.

On the other hand on applying the t-test for Agra, a historical city[9] of India (roadside soil data 12.9 μg g^{-1}), the data of two places does not differ significantly at 5 % level of significance. This indicates almost equal level of PAHs contamination at two places.

Comparison of Jalandhar city average (16.38 μg g^{-1}) of 16 PAHs with selected literature data, it was found toward higher side to that of many other cities of the world such as Kota Bharu Malaysia, 1.45 μg g^{-1} [15], Australia, 3.30 μg g^{-1} [10], Delhi airport soil, 4.43 μg g^{-1} [14], Agra, India, 12.9 μg g^{-1} [9], but lower than few other city of the world such as USA, 58.68 μg g^{-1} [16].

Conclusion

This is the first comprehensive information on PAH spatial distribution pattern at different distances (1, 2 and 3 m) from roadside soil in a fast developing city of India. This monograph studies of roadside soil contamination with PAHs and found that 1 m distance from roadside is highly contaminated with many carcinogenic PAHs. The behaviour was because of close intercepts (building, plants and shops) at these intercepts whereas, in the case of average population and traffic density intercepts behaviour was somewhat different.

Among five major (average within 3 m) PAHs, IP and B(a)P were the individual PAH present in highest concentration (ranging between 30.09-169.62 and 16.02-159.51 μg g^{-1}) at all intercepts. Some of the PAHs were found to settle down at closer distance to the roadside as indicated by their absence at longer distance in soil at many intercepts.

It was observed that carcinogenic PAHs (4-6 rings) were in higher concentration (85 %) than non-carcinogenic PAHs (2-3 ring, 15 %) at most intercepts. City average (for all ten intercepts within 3 m) of non-carcinogenic and carcinogenic PAHs was 4.86 and 31.39 μg g^{-1} (ratio 1:6.45), which was quite high could be toxic to human. It was also concluded from this study that even developing cities are showing equal or even higher concentration of carcinogenic PAHs than metro cities, which is a dangerous sign toward human exposure. The study could be of great significance for the planers while considering environmental remedial measures.

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